Behavior and distribution of plastic debris in the urban river in Mongolia

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論文名

Behavior and distribution of plastic debris in the urban river in Mongolia

（モンゴルの都市河川におけるプラスチックごみの動態と分布）

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ABSTRACT

Remarkably high utility of plastics has promoted the drastic increase of plastic production and consumption over the last several decades, resulting in generation of large amount of plastic solid waste and further its accumulation in the environment. Although the majority of plastic debris in the marine environment derived from land-based sources, the transportation processes through river systems are unclear but important. Since plastic debris pose a threat to aquatic ecosystem, the current thesis aimed to identify behavior and distribution of environmental plastics in the Selenga River system, particularly along the Tuul River including Ulaanbaatar City of Mongolia. Plastic debris in the aquatic environment affected by plastic production and its waste generation was reviewed and its consequences relating to their behavior in the environment were pointed out in Chapter 1.

There are no universally accepted methods to determine plastic debris. Considering many adopted methods for sampling, sample preparation and analyses of environmental plastics, validation and standardization of the most appropriate methods achieve high separation efficiency of plastic debris from the environmental matrix. The suitable methodological approaches for sampling, sample preparation and analysis of plastics in river environments were proposed and discussed in Chapter 2. Moreover, river shores and bottom sediments were selected as important sampling spots for the river environment to obtain comprehensive data of distribution pattern and degradation processes of environmental plastics based on hydrological and geographical backgrounds of the Selenga River and Tuul River systems. Nevertheless, from the methodological viewpoint, the carbonyl index was considered as a good indicator to identify plastic origins and surface deteriorations simultaneously.

In chapter 3, abundance and distribution of visible plastic debris on the shores of Selenga River system were addressed to reveal the distribution pattern and fragmentation process of plastics in the river environment. Diversity in abundance and characteristics of the plastic debris on the river shore was probably reflected by multiple pathways of plastic debris in the river catchment. On-site fragmentation of plastic debris has been speculated in the foam and film morphotypes of plastics due to freeze-thaw and dry-wet cycles. Despite of occurrence of different morphotypes of mega-, macro-, meso- and micro- plastics on the river shores, micro sized polystyrene foam (PSF) have been considered as distinctive plastic debris in both the Selenga River system and the Tuul River system due to its predominant production and consumption in Mongolia.

In Chapter 4, the major plastic debris of PSF was focused to characterize their degradation status and formed aggregates with invisible microplastics (MPs) in the urban river environment. Fragmentation of PSFs evaluated from the composition of size fractions of PSFs revealed the behavior and fate of plastic debris after their release into the environment. Although surface status of PSFs indicates the different residential time in the environment, fragmentation and biofilm coverage influenced the surface status expressed by carbonyl index (CI) which is used for estimation of photodegradation. The range of CI in the size fractions of PSFs was large in the micro sized PSF, indicating that frequent fragmentation of PSFs makes different surface status on micro sized PSFs. Number of invisible MPs adhered onto the surface of PSF were relatively high in the micro sized PSF fraction. PSFs are one of the carriers of invisible MPs in the river environment. These aggregates could be found in any aquatic environments due to their ubiquitous distribution in marine and freshwater environments.

In Chapter 5, settled MPs in the river bottom sediment was targeted to understand deposition of plastics and their characteristics in the bottom sediment in the Tuul river. The river sediment contained plenty of micro-fibers probably due to mixing the effluent from the wastewater treatment plant (WWTP). The major plastic composition detected as settled plastics was different from that on the river shore, where PSF was predominant. The deposited plastics such as polyethylene and polypropylene which have lower specific density than that of water demonstrate that deposition of the light plastics was occurred as a result of surface changes of plastics through not only biofouling but also formation of aggregates with suspended sediments in the river environment. Although the mechanisms of aggregation consisting of plastic debris and minerals was not clear, the MPs can interact with suspended sediments because of their high specific surface area.

The behavior of plastic debris in the urban river environment indicates that plastic consumption depending on population density in urban areas is a significant controlling factor on the distribution of plastic debris along the river. Considering to plastic fragmentation into micro- and nano-scale sizes, number of plastic debris affecting to ingestion of aquatic organisms increase in bioaccumulation of various aquatic organisms. The process of fragmentation and the behavior of small sized fragments in the river environment is necessary to take into account by long-term monitoring followed by modeling using the monitoring data. Finally, present study highlighted the behavior and distribution of PSFs and the status of PSFs as aggregates in the river environment.

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ABBREVIATIONS AND ACRONYMS

EU	European Union
FTIR	Fourier Transform Infrared Spectroscopy
HCA	Hierarchical Cluster Analysis
IR	Infrared
MP	Microplastic
MSW	Municipal Solid Waste
NOAA	National Oceanic and Atmospheric Administration
PCA	Principal Component Analysis
POPs	Persistent Organic Pollutants
PSF	Polystyrene Foam
SI	The International System of Units
SD	Standard Deviation
SE	Standard Error
UK	United Kingdom
USA	United States of America
UV	Ultraviolet
WWTP	Wastewater Treatment Plant

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CHAPTER 1

Introduction to plastic debris in aquatic systems

1.1 Background

In these several decades, anthropogenic litter has significantly increased in marine and freshwater environments and has become a serious environmental issue in the world. Numerous scientific reports have criticized anthropogenic litter, particularly plastic debris in aquatic environments, which have affected organisms from the various parts of the world (Ali and Shams, 2015; Hermabessiere et al., 2017). Plastic products are chemically synthesized from fossil hydrocarbons and most of them are non-biodegradable. Drastic increases of plastic manufacture and plastic consumption in our daily life cause generation of large amount of plastic solid waste and further accumulation in environment. Following the worldwide plastic production (322 million tonnes of in 2015, PlasticsEurope, 2016), world ocean received 4.8 to 12.7 million tonnes of plastic waste in 2015 mainly by river transport (Jambeck et al., 2015). The recent estimation states that both statics of plastic production and waste generation with their release as plastic litter will continuously increase in the future. Associating with non-biodegradable characteristic, chemical and physical properties of environmental waste plastics are altered through mechanical and physical breakdown, thermal and photochemical degradation, biofouling and aggregation (Zhang, 2017). Modification of plastic properties under the environmental condition affects abundance and behavior of plastic debris which arise various concerns in the environment.

Plastics are relatively persistent, and this property makes plastic pervasive as an environmental pollutant (Horton et al., 2017b). Plastic litter has been ubiquitously observed in freshwater, marine, estuarine and coastal environments (Andrady, 2011). Moreover, it has mixed together and forming ‘plastiglomerates’ with volcanic materials, sediment and organic materials (Corcoran et al., 2014). Generally, molten plastics had infilled vesicles in volcanic rocks, thereby forming plastic amygdales (Fig. 1.1). Formed ‘plastiglomerates’ have a potential to become preserved in the fossil record. Recently, the ‘plastiglomerates’ or ‘plasticrusts’ (Gestoso et al., 2019) are getting more broadly discovered in many coastal areas (Malizia and Monmany-Garzia, 2019). Consequently, these pollutants can be a distinct marker of anthropogenic activity. According to the geologic timescale, we are currently

living in the Holocene epoch. Since anthropogenic activity with large impacts started to dominate the most fundamental processes on earth, some geoscientists proposed the term “Anthropocene” to represent the period of time between the latter half of the 18th century and the present day (Crutzen and Stoermer, 2000). Recent studies also discussed that the plastic materials have a strong potential to be a global marker horizon in the Anthropocene due to their ubiquitous distribution and influences on the planet's biota through ecological cascades including human-well-beings (Malizia and Monmany-Garzia, 2019; Zalasiewicz et al., 2016).

1.2 Plastic production, waste generation and legislation in the world

Plastic production

Plastics are incredibly versatile, durable and inexpensive which attributes a wide range of applications (Frias and Nash, 2019). Based on its unique properties, such as durability, resistance, light-weight, electrical insulation and strength, a mass production has started from 1950’s. Numerous applications in many industrial sectors of plastics have been achieved, resulting in drastic increase of plastic production from 1.5 million tons in 1950’s to approximately 350 million metric tons in 2017 (Fig. 1.2, Geyer et al., 2017). The term of plastics can be applied to a wide range of synthetic or semi-synthetic polymers. Thermoplastics and thermosetting plastics are the major classification of plastic materials based on their thermal property (PlasticsEurope, 2018). Thermoplastics are reversible plastics that can be remoulded when heated or cooled, whereas, thermosetting polymers undergo a chemical change into a particular shape and cannot modified again due to crossed linkage between chain polymer structures when they are heated. Widely used single-use plastic materials such as polypropylene, low and linear low-density polyethylene, high-density polyethylene, polyvinyl chloride and polystyrene are dominant thermoplastics in the world plastic market (Geyer et al., 2017). Polyurethane, polyester, epoxy resin and acrylic resins are other frequently used thermosetting plastics in the world. The consumption of plastics is dominated by packaging (44.8%), building and construction (18.8%), and institutional products (11.9%) (Geyer et al., 2017). Single-use disposable applications such as packaging, agricultural films and disposable consumer items have occupied approximately half of plastic production (Hopewell et al., 2009).

Management for plastic solid waste

Plastic products released to environments as the huge amount of plastic wastes which ends up as a litter and/or municipal solid waste (MSW). Plastic solid waste is produced on a massive volume worldwide and generation of plastic wastes reaches to the 150 million tons per year globally (Singh et al., 2017). Additionally, a model calculation of municipal plastic solid waste has estimated between 60 and 99 (mid-point: 80) million metric tonnes of plastics which were inadequately disposed globally into the environment during 2015 (Lebreton and Andrady, 2019). Figure 1.3 shown the global mismanaged plastic waste generation in 2015 (Lebreton and Andrady, 2019). Since approximately 10% of global MSW is occupied by plastic solid waste, it is important to manage these plastics by proper waste management (Mwanza and Mbohwa, 2017). Recycling, landfilling and incineration methods have been subjected to manage MSW in the world (Tripathi et al., 2016).

Recycling is the best choice for plastic solid waste management through several steps of industrial processing. With technological advancement in modern industries, all types of plastics can be recycled by various technologies (Singh et al., 2017). However, the heterogeneity of plastic products, types of additives and differences between manufacturing processes make it difficult to recycle all plastics in many cases (Hopewell et al., 2009). On the other hand, repetition of recycling procedures makes plastic materials weak and lose their strength which is required for the next recycle of plastic solid waste management.

Electricity regeneration using heat through incineration of plastics as a waste management has become a popular option as energy regeneration in the world. Reduction of waste plastic volume reaching to 90 − 99% occurs during the incineration, resulting in reduction in volume of final wastes for landfilling (Al-Salem et al., 2009). Although the energy recovery is collected by burning of the plastic wastes, the process accompanies releases of highly toxic chemicals such as polycyclic aromatic hydrocarbons, volatile organic compounds, SOx and particles into the air (Al-Salem et al., 2009; Antelava et al., 2019). For this reason, it is necessary to appropriately control the emissions during the incineration process. Although energy recovery is one of the important ways to solve increasing trend of plastic solid waste, the associated cost for the management of plastic solid waste leads any countries and communities to choose another option (Antelava et al., 2019).

Landfilling is widely applied method to manage not only plastic solid waste but also MSW in the world. Geyer et al., (2017) have estimated that 78% of all produced plastic waste from 1950 – 2015 has discarded into landfills, suggesting that a large amount of buried

plastic wastes might have severe effect to the environment. Additionally, landfilling with mixed waste is the least environmentally friendly option, despite of least cost and one of the traditional methods of waste management in the world (Horodytska et al., 2018).

Legislation of plastic wastes in the world

Many countries and communities have established policies and regulations for plastic waste management, aiming to reduce amount of waste through reuse, recycling and recovery of various plastic solid waste (Singh and Sharma, 2016). For example, European Union (EU) has encouraged the post-consumer recycling through the policy of EU Directive on packaging and packaging waste (94/62/EC). Proper treatment has reported not only European countries but also USA, India and other counties (Wagner, 2017). Many countries implemented a ban on plastic bag due to its large production and the single-use management.

Although worldwide legislations have been developed for many plastic goods based on their utility and environmental burden, cost-effectiveness, advanced ways to reduce plastic production and waste generation should be considered taking the terrestrial and marine pollution into account (Auta et al., 2017). Furthermore, estimating global plastic waste generation and recycling rate are required as annual national data for waste management practices in terms of activities for reduction of plastic waste and improvement of regulations to avoid uncontrolled release of plastic solid waste into the environment. Although research reports provide the data related to plastic waste which collected during the research period, many countries especially low and lower middle-income countries are lacking consistent data of plastic wastes (Singh and Sharma, 2016). Therefore, the national statistics associated with waste, a standardization of terminology and consistent activity are necessary to improve data quality for this sector.

1.3 Physical and chemical properties of environmental plastics

Environmental plastics have influenced by various mechanical and physical forces in the outdoor condition which leads to surface changes (e.g., weathering, embrittlement, degradation, fragmentation) on plastic debris. The degradation processes of all types of plastic materials firstly are induced by ultraviolet (UV) light (photooxidation), resulting in weathering of the surface displayed by surface roughness, variations in topography of the surface, followed by holes and micro-cracks on the surface (Syranidou et al., 2019). Due to the photoinduced processes, the polymer usually changes its appearance (surface gloss and

color) followed later by changes in mechanical properties (strength, strain and flexibility) and increasing their fragmentation in the environment.

According to broad physical and chemical properties of plastics, environmental plastic debris is classified by size category, morphotypes and polymer origins. Plastic debris with the wide range of sizes from the microscopic to metric size have been found in all aquatic environments from coastal areas, terrestrial and ocean such as the Antarctic, the Arctic and the Southern Oceans (Barnes et al., 2010; Browne et al., 2011). Megaplastic (>100 mm), macroplastic (100 − 20 mm), mesoplastic (20 − 5 mm) and microplastic (MP; <5 mm) are the primarily used size category of plastic debris (Barnes et al., 2009; Horton et al., 2017b). Additionally, nanoplastic size ranges between 1 and 100 nm (EFSA, 2016). The size category of any pieces of plastic was termed according to above mentioned size ranges along its longest dimension in order to characterize the behavior of plastics in the environment.

In terms of morphotypes, plastics have been classified into five groups: foams, films, fibers, fragments and pellets (Rezania et al., 2018). Expanded and extruded polystyrene (e.g. styrofoam, spongy plastic) like as insulation materials are categorized into the foam type of plastics (Davis and Murphy, 2015). Films are flexible and very thin plastics which are likely to be derived from packing materials (Horton et al., 2017a). Synthetic fishing line, ropes or clothes are included in the fiber types, whereas microbeads, pre-produced plastic nurdles and resins are considered as the pellet types (Anderson et al., 2016b). Hard plastics that are generally unrecognizable plastics are included in the type of fragments (Carson et al., 2013). An example of each morphotype have shown in the Figure 1.4 (Free et al., 2014). Identification of plastic morphotypes provides beneficial information relating to polymer types and origins. For example, films usually originated from packaging materials, which generally made from polyethylene thermoplastic and its derivatives. The polystyrene, polyurethane and acrylonitrile butadiene styrene are the major constituents of foams, which are mainly used for packages, food containers and insulation materials. Usually, polymer origins can be identified from spectra using instrumental analyses (such as Fourier transform infrared (FTIR) spectroscopy, Ramen spectroscopy). Various types of plastics such as polyethylene, polypropylene, polystyrene, polyurethane, polyvinylchloride, etc., are able to identify referring with standard samples (Eerkes-Medrano et al., 2015; Hidalgo-Ruz et al., 2012; Van Cauwenberghe et al., 2015).

Furthermore, aged plastics demonstrate an increase in oxygenated moieties such as carboxylic acids, aldehydes, hydroperoxides, ketones and alcohol groups in the plastics as a result of prolonged exposure to UV radiation (Barbeş et al., 2014). Carbonyl functional

groups can be used to identify the degradation stage of environmental plastics (Andrady, 2011). Chemical changes of a polymer material which associated with aging can be estimated by carbonyl index. Carbonyl index is probably the most commonly used indicator to measure the chemical oxidation of plastic materials using a photodegradation approaches relating to the functional properties of plastic materials (Rouillon et al., 2016). The ratio of carbonyl moieties absorbance and reference moieties absorbance is estimated as a carbonyl index in the aged plastics (Botelho et al., 2004; Mylläri et al., 2015). The carbonyl index is calculated using equation (*1.1*):

$$Carbonyl\ index = \frac{A_{-CO-}}{A_{-CH_2-}} \qquad (1.1)$$

where A_{-CO-} is the absorption from carbonyl group observed at suitable wavenumber in each polymer (e.g. 1456 cm^{-1} or 720 cm^{-1} and 730 cm^{-1} for polyethylene; Andrady, 2017) and A_{ref} is the absorption from reference group (usually methylene group) observed at suitable wavenumber in each polymer (e.g. 2850 cm^{-1} or 2920 cm^{-1} and 1880 – 1910 cm^{-1}; Shi et al., 2019; Tocháček and Vrátníčková, 2014).

Physical and chemical properties of small sized plastics such as MPs and nanoplastics have been challenged considering their technological limitations to identify and characterize them (Filgueiras et al., 2019). However, size distribution, shapes and colors of MPs in marine and fresh waters have received particular attentions to evaluate their behavior in environments. Microplastics are broadly classified as primary or secondary sourced MPs based on their form that found in the environment. Primary MPs are manufactured to be micron size and it includes resin pellets and microbeads derived from personal body care products (Anderson et al., 2016a). Secondary MPs are formed by mechanical breakdown or fragmentation of larger plastics (Eerkes-Medrano et al., 2015). Massive amount of secondary MPs is distributed in the marine and terrestrial environment (Barnes et al., 2009; Horton et al., 2017b). It has been difficult to estimate input of secondary MPs from terrestrial to aquatic environment due to unexpected number of daughter fragments. Therefore, given their variety of chemical and physical properties, it is more complicated to speculate fate of MPs in the aquatic and terrestrial environments.

1.4 Plastic debris in the environment

1.4.1 Marine plastic debris

Studies on occurrence and accumulation of anthropogenic litter in marine environment usually find the plastic debris to be predominant component of the mix of debris (Galgani et al., 2015). Plastic debris is reaching to 60 − 80% of the world's litter (Retama et al., 2016), and about 90% of floating debris is considered to be plastics in the world oceans (Moore, 2008). Almost half of world's population concentrated within 60 km from the seacoast and 75% of all large cities located on the coast (GESAMP, 2015). Given the attention to the large amount of plastic debris in marine environment, it is reasonable to assume direct and indirect inputs of plastic debris from urbanized areas to aquatic environment (Fig. 1.5). The plastic release from coastal countries was estimated that 1.7 to 4.6% of the total plastic waste generated in coastal 192 countries due to mismanagement leading to the ocean in 2010 (Jambeck et al., 2015).

Gyres (North Pacific, South Pacific, North Atlantic, South Atlantic, Indian Ocean) are particular hotspots for plastic debris accumulation in the world ocean (Fig. 1.6) (Maximenko et al., 2012). Prediction models of plastic accumulation including spatial and temporal distribution of plastic debris illustrated that high concentration of plastic debris concentrated in the North Atlantic Ocean gyre as a result of the highest coastal population (Enders et al., 2015; Lebreton et al., 2018). Recently notably high concentration of MPs reaching to 32.76 particles m^3 has been recorded in the North Pacific subtropical gyre (Goldstein and Goodwin, 2013).

Prediction and estimation of plastic debris in the world ocean have been conducted taking into account a significant consideration in consequence of largely distributed plastics and their behavior regarding to the interaction with wildlife. Using the reported research database and world statistics, a recent estimation showed that 5.25 trillion plastic particles (weighing 268,940 tons) have floated on the world ocean surface in 2010 (Eriksen et al., 2014). On the other hand, the recent research shows another calculation which was estimated the concentration of floating small plastic debris with 15 to 51 trillion particles, that was consistent to weight ranging between 93 and 236 thousand metric tons in 2014 (van Sebille et al., 2015). The estimation in 2014 is far more than that in 2010, suggesting that only approximately 1% of global plastic waste entered into the ocean in the year 2010. Koelmans et al., (2017) estimated the fragmentation and vertical transport of oceanic plastic in 2016.

The simulation showed that the 99.8% of plastic debris reaching to the ocean since 1950 had settled below the ocean surface layer until 2016, which was based on abundance of micro- and macro-plastics in the ocean surface layer. Additionally, 0.309 million tons of plastics (83.7% - macro, 13.8% - micro and 2.5% - nano plastics) have floated in the ocean surface layer in 2016.

Plastics have been produced just from 1950's and it was relatively new materials. Within the production and consumption during about 60 years, plastics have been found everywhere in the marine environment (Galgani et al., 2015; Thompson et al., 2004). Marine scientists largely contributed to develop database of abundance and distribution of floating, submerged and deposited plastic debris based on consequences of environmental cleaning campaign for the solution to aesthetic problem in the beach and ocean surface water, and to remove entanglement of aquatic organisms. However, it has been gradually extended their research fields for surveys of environmental plastics with different purposes from interdisciplinary scientific viewpoints. One of the important viewpoints is plastic loads in terrestrial environment and estimation of transporting plastics from terrestrial environment to open water systems through river pathway, which recently appeared in the published scientific works.

1.4.2 Freshwater plastic debris

The distribution and occurrence of plastic debris in terrestrial freshwater systems was unknown as those in marine systems until recently. Given the attention to the receiver and transporter of plastic debris in aquatic environments, many studies started to highlight the occurrence and abundance of plastics in the terrestrial freshwater systems (Eerkes-Medrano et al., 2015). Additionally, studies of coastal environments have pointed out that densely populated urban environments are important sources of MPs to river outlets. The published research works illustrate that concentrations of plastics, especially MPs, in different freshwater compartments were largely varied in different sampling locations. Large rivers and lakes have reported the various concentration of plastic in the water (Table 1.1), bottom sediment and on the shore (Morritt et al., 2014; Wang et al., 2017b; Zhang et al., 2015).

The study of River Thames in United Kingdom (UK) has targeted submerged plastics in the river and reported the 8490 plastics with dominance of sanitary products over a three-month monitoring (Morritt et al., 2014). In China, there are several studies which highlighted the MPs in freshwaters. The 20 urban lakes and urban reaches of the Hanjiang River and

Yangtze River of Wuhan have been investigated by Wang et al., (2017c). The abundance of MPs was ranged from 1,660.0 ± 639.1 to 8,925 ± 1591 number m^{-3} for the studied 20 waterbodies. Additionally, the study found the negative correlation between MP concentrations and the distance from the urban center and noted the potential sources of higher population densities and anthropogenic activities. The Wuhan Municipal Government was assumed to be the urban center in the study. Furthermore, the MPs in water and sediments has been detected in Pearl River along Guangzhou City ranging from 379 to 7924 items·m^{-3} and 80 − 9597 items·kg^{-1}, respectively (Lin et al., 2018). Although multiple sourced micro-fibers dominated both in water and sediment samples of Pearl River, the study noted that effluent from Wastewater Treatment Plants (WWTPs) couldn't be neglected. Saigon River in Vietnam have received large amount of macro and micro-plastics from surrounding environments which reported by Lahens et al., (2018). Land-based plastic debris have been accounted from 0.96 to 19.91 g $inhabitant^{-1}$ d^{-1}, namely 350 to 7,270 g $inhabitant^{-1}$ yr^{-1} in the Saigon River. Various number of MPs have been determined in the 29 Japanese rivers (Kataoka et al., 2019). The mean abundance was 1.6 ± 2.3 pieces m^{-3} in the river water and the results demonstrated that not only high population density area and urbanization but also river inflow process have affected the concentration of MPs in the rivers (Kataoka et al., 2019). Considering the river plastic flux, researchers focused on the MP flux from rivers to open water system. The MP flux was measured from 15,520 pieces to 4,721,709 pieces per day in nine rivers in Illinois, United States of America (USA) (McCormick et al., 2016).

Recently, the number of MPs in the river bottom sediment have received particular attention from the researchers due to the bottom sediment can be considered as a major sink of plastics in the terrestrial environment. In the study on sediments of River Thames, the abundances of MPs have varied between 18.5 ± 4.2 and 66 ± 7.7 particles/100 g (Horton et al., 2017a). The highest value of MPs in sediment might be derived from the thermoplastic which was used as road-surface marking paints through surface runoff in the urban area. In the Atoyac River in Mexico, colored (51%) and white (49%) MPs were predominantly determined in sediments with total number of 4500 ± 702.2 items kg^{-1} (Shruti et al., 2019). The data from Atoyac River indicated anthropogenic stress such as industries, population density and number of WWTP which can release MPs with the various concentration into the sediments.

Eriksen et al., (2013) have addressed the MP pollution in the surface water of Laurentian Great Lakes in the US. Average abundance of MPs was approximately 43,000 particles km^{-2}.

However, some sampling sites in Lake Erie recorded extraordinarily high number of plastic particles released from industrial areas mainly used for coal burning industry and sewage effluent from WWTP. Remotely accessed mountain lake, Hovsgol, in Mongolia, have been investigated about the distribution of plastic debris at the shoreline and in the pelagic region (Free et al., 2014). Surprisingly large number of MPs has been detected in the remote Hovsgol lake in Mongolia. Comparing to Lakes Huron and Superior in the Laurentian Great Lakes, Hovsgol lake was heavily polluted by MPs with the average concentration of 20,264 particles km^{-2}. Without waste management, household and recreation resort wastes were discarded into surrounding illegal dumping sites, leading to occurrence of plastic debris at the lake shore and in the pelagic zone of the Hovsgol lake.

Although the number of studies on plastic debris in the aquatic environment, particularly on river systems, has increased to evaluate the MP behavior affecting to the marine environment, the dynamics of environmental plastics in any freshwater compartments as well as their degradation processes are still unknown (Horton et al., 2017b). Moreover, release of plastic debris from inland areas is currently unclear because of lack of data. Specifically, information of MP transportation in the terrestrial ecosystems through surface runoff and stream flows is missing. Therefore, behavior of plastic debris is required to understand the processes associated with plastic debris in the river environment near high population density area.

1.4.3 Consequences of plastics in the aquatic environment

In recent years, concerns on plastic debris in the aquatic systems have expanded to various organisms, resulting in injury of them through plastic ingestion (Cole et al., 2013; Hall et al., 2015), entanglement and ghost fishing (Brown et al., 2005; Gregory, 2009), adsorption of heavy metals and toxic organic compounds (Hüffer et al., 2018; Liu et al., 2019b), and vectoring adsorbed pollutants (Didier et al., 2017; Kirstein et al., 2016). MP ingestion by various organisms have been widely reported in the marine and freshwater environments (Cole et al., 2013; Setälä et al., 2014). Additionally, translocating MPs from digestive tract to other tissue of organism and transfer potential of MPs in the aquatic food web have been received as an important concern from the viewpoint of bioavailability of plastics (Browne et al., 2013; Setälä et al., 2014). Zooplankton and mysid shrimps are used to understand food chain transfer of MPs by experimental analysis (Setälä et al., 2014). The study was based on feeding the mysid shrimps by zooplankton which was labelled with

ingested microspheres. After the incubation for 3 hours, the result demonstrates the MPs transferred from one trophic level (mesozooplankton) to a higher level (macrozooplankton). Many researchers handled the laboratory methods to speculate MP ingestion, stress by feeding, decrease in reproductivity, response from body after the ingestion and vectoring other pollutants into organs. However, the precise mechanisms of MPs translocation in organisms and transferring into food chain still remain unknown.

Importantly, there are numerous toxic chemicals and organic pollutants existed in the aquatic environment, and plastics have a potential to adsorb those pollutants from the surrounding environment (Andrady, 2011; Wang et al., 2016). Plastic debris, particularly with the high specific surface area of MPs, are susceptible to contamination by different waterborne-pollutants including aqueous metals (Maršić-Lučić et al., 2018; Mohsen et al., 2019), endocrine disrupting chemicals (Zhang et al., 2018b) and persistent organic pollutants (POPs) also referred as hydrophobic organic contaminants (Cole et al., 2011). Furthermore, MPs might potentially transfer adsorbed chemical pollutants from the environment to aquatic organisms (Zhang et al., 2018b). At the same time, plastic additives may represent adverse impacts on aquatic ecosystems. There are various types of chemical additives widely used in manufacturing processes of plastic materials. For example, they are stabilizer, antioxidant, flame/fire retardants, heat stabilizers, UV stabilizers, colorants, cross-linking agents, impact modifier, blowing agents, fillers and antistatic (Teuten et al., 2009). Polybrominated diphenyl ethers, nonylphenol and triclosan are mainly used for resistance to heat, oxidative damage and microbial degradation, respectively (Andrady, 2011). During the environmental travel, plastics gradually lose their physical properties, leading to leach out of hazardous chemical additives from the plastics followed by their accumulation in the environment (Barnes et al., 2009). As a consequence of plastic accumulation and fragmentation in the oceans, plastic additives could represent an increasing ecotoxicological risk for marine organisms (Hermabessiere et al., 2017).

Although more accurate assessments in terms of realistic exposures will be required to the future prediction of pollutants, based on laboratory experiment and model approaches, aged plastics especially MPs have posed serous concerns relating to leaching of chemical additives and adsorption of other pollutants such as heavy metals and POPs.

1.5 Social issues of wastes in Mongolia and plastics in the river system

Mongolia is a landlocked country located in the central part of Asia between China and Russian Siberia (Fig. 1.7). The population of Mongolia with 1,564,116 km^2 land is around 3 million which indicates one of the most sparsely populated countries in the world. Population and economic growth with urbanization accompanied by modern lifestyle have been accelerated in most of developing countries including Mongolia after 1990's democratization. As a result, solid waste generation have rapidly increased and the composition of MSW has become more diverse. In Mongolia, a variety types of solid wastes released from industries, agriculture and daily life are mainly managed by a landfill method (Fig. 1.8 a-b). Ulaanbaatar city, the capital of Mongolia, is facing the waste related issues due to lack of proper regulations.

The generation of MSW has increased with its diversity of origins year by year. In 2015 and 2018, 2.3 and 2.9 million tons of waste were disposed into 371 landfill sites in Mongolia, respectively (Environmental Information Center 2015). The most of generated wastes (approximately 90%) were transported to the landfill sites and approximately 10% of the waste have been lost in the surrounding environment during transportation and/or illegal deposition (Delgermaa and Matsumoto, 2016; JICA, 2012). Due to increase of illegal deposition in the public spaces and related environmental pollution, the government is seeking a new MSW management system (Toshiki et al., 2015). Japan International Cooperation Agency suggested the "Solid Waste Management Plan for Ulaanbaatar City, Mongolia" based on a separation technique for paper, plastics and other wastes to produce refuse derived fuel (JICA, 2012). The aim of MSW management is reuse of wastes as a resource of fuel to produce heat and to reduce the landfill volumes. However, the suggested waste management is still under the procedure to construct recycling industry.

Officially, there are several landfills located in the towns or villages where mainly lies in the upland area (Byamba and Ishikawa, 2017). The three main landfill sites in Ulaanbaatar weekly take approximately 18,000 tons of wastes, which include polyethylene terephthalate bottles and other plastic wastes with the rate of 2% of the whole of wastes (Mongolian National Recycling Association 2018). Due to lack of tracks to collect the MSW from residential areas and insufficient road infrastructure to reach some households in hills and mountainous areas, the number of illegal dumping sites have increased and waste dispersion from landfill and illegal dumping sites are particular sources of plastic debris in the environment. Scattering of wastes from small dumping sites by wind is also common. To

clean up scattered wastes, social volunteers and city official workers started to collect disposed waste along the roads, public spaces and riversides in urbanized areas (Fig. 1.8c, Tuul River Basin Authority 2017). For example, the Tuul riverside in the Ulaanbaatar City area was cleaned up 13 times in a year and 159.4 tons of waste were collected in the 67 km^2 area of the river catchment by volunteers in 2017. Clean up activities usually focus on macro-sized plastic debris and most collected waste includes polyethylene terephthalate bottles and plastic polyethylene sheets.

The largest river system of the Selenga River (Fig. 1.9), is a transboundary water system starting from the Khanghai Mountains in Mongolia finally flowing into Lake Baikal, which is the world's largest freshwater reservoir (Baljinnyam et al., 2014). One of the main tributaries in the Selenga River system is the Tuul River which is running through the city center of Ulaanbaatar. Residential areas and low levels of sewage water systems have been recognized as a major pollution source of the river water (Itoh et al., 2011). In some residential areas in the Tuul River basin, household waste and sewage water have been directly dumped to the nearest illegal dumping sites located on hills, on yards and alongside roads and waterways (Delgermaa and Matsumoto, 2016). The tributaries are also subjected to various types of pollution by mining activities, agriculture, pastureland degradation, and WWTP in their watersheds (Batbayar et al., 2017; Nadmitov et al., 2015). Broader and intensive inspection and strict control of the illegal disposal of waste are now considered along the urban rivers in Mongolia. Low-density and smaller-sized plastics, which are easily blown away by wind, are trapped by plants on the river shores and are redistributed by river flow during the snowmelt and rainy seasons when the water level and flow rate are high. Recently particular attention has been paid for MPs due to their dynamic behavior as plastic particles in water systems.

1.6 Aim of the thesis

Among the multiple human pressures on aquatic ecosystems, the accumulation of plastic debris is one of the most obvious concerns despite of the least research achievements. Although plastic materials have remarkable societal benefits, there are drawbacks to our 'plastic ages' (Thompson et al., 2009). Global monitoring of marine plastic debris has succeeded to understand the fate, distribution and potential risks of plastics, while behavior of plastic debris in the terrestrial environment including river ecosystems from its occurrence to release into marine environment is still largely unknown.

Given our limited knowledge about plastic debris in the urban river environment, current thesis aimed to understand the fate and behavior of plastic debris in the terrestrial river environment. The objectives of this thesis are as follows:

1) to review various methodologies for plastic sampling, separation, determination in environmental systems, especially focusing to riverine environment;
2) to elucidate occurrence and fragmentation of plastic debris on river shores of the Selenga River system;
3) to recognize the degradation status of polystyrene foam (PSF) and interaction between PSF and other MPs in the urban river in Ulaanbaatar, and the amount and properties of adhered MPs on the surface of PSFs;
4) to understand dynamics of plastics in the freshwater system taking into account a sedimentation process and identification of possible mechanism of deposition of MPs in the bottom of the urban river ecosystem;
5) finally, a conclusion will be addressed the behavior and fate of plastic debris in the urban river environment. Ultimately, outlook of environmental plastic research from the perspectives of current study will be proposed in order to understand current issues and avoid future problems relating to large amount of synthetic plastic consumption.

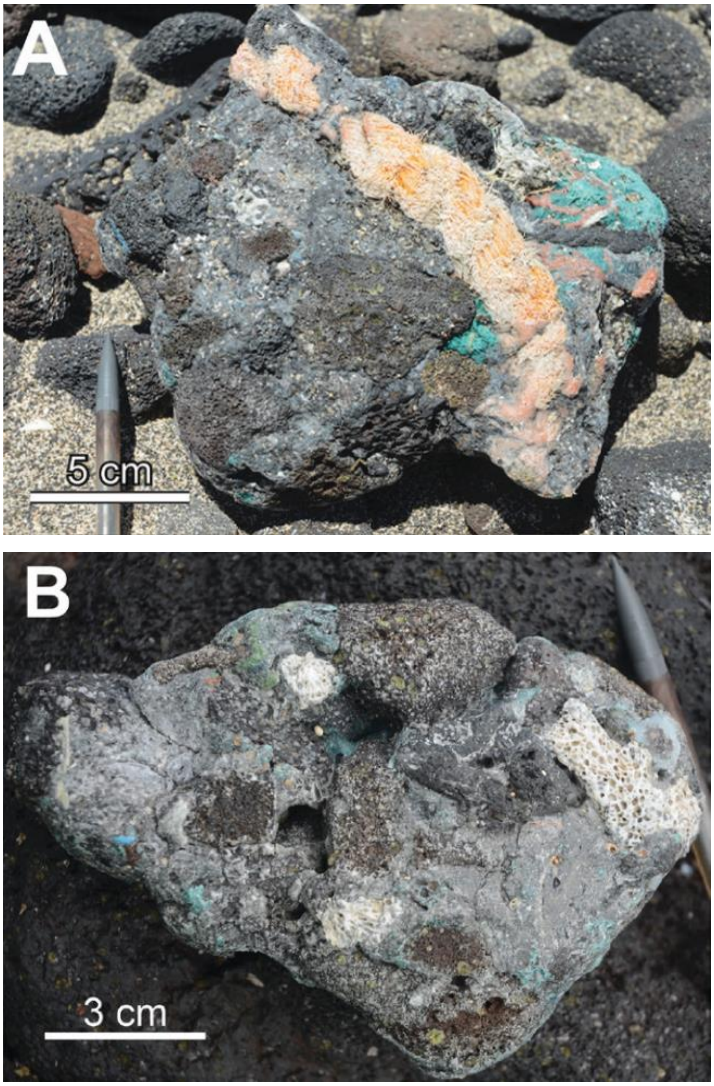

Fig. 1.1 Photographs of clastic plastiglomerate on Kamilo Beach (Corcoran et al., 2014). (A) Subrounded fragment containing basalt clasts, molten plastic, yellow rope, and green and red netting; (B) Clastic plastiglomerate containing molten plastic and basalt and coral fragments.

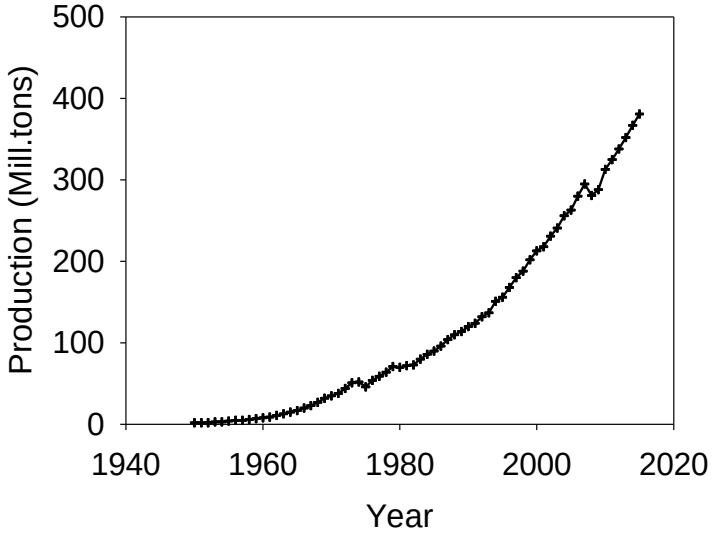

Fig. 1.2 Annual global plastics production in millions of metric tons (Geyer et al., 2017).

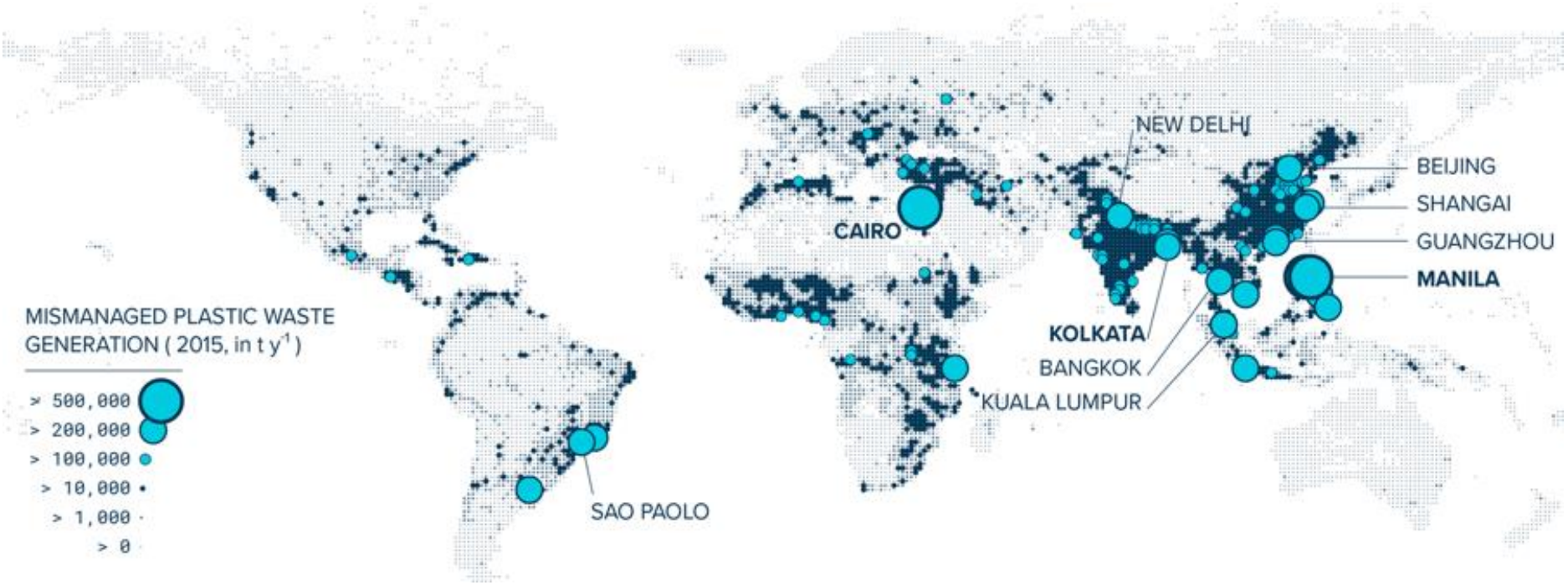

Fig. 1.3 Global mismanaged plastic waste generation in 2015. Plastic waste generation is computed globally on a 30 by 30 arc seconds resolution. The map reflects geographical heterogeneity of plastics based on population and gross domestic product distributions. National data on waste management reported in each country (Waste Atlas, 2016) is cited to estimate the mismanaged fraction at local scale. The 10 largest producing urban centers are labelled on the map with Manila, Cairo and Kolkata as the leading agglomerations (See details in Lebreton and Andrady, (2019)).

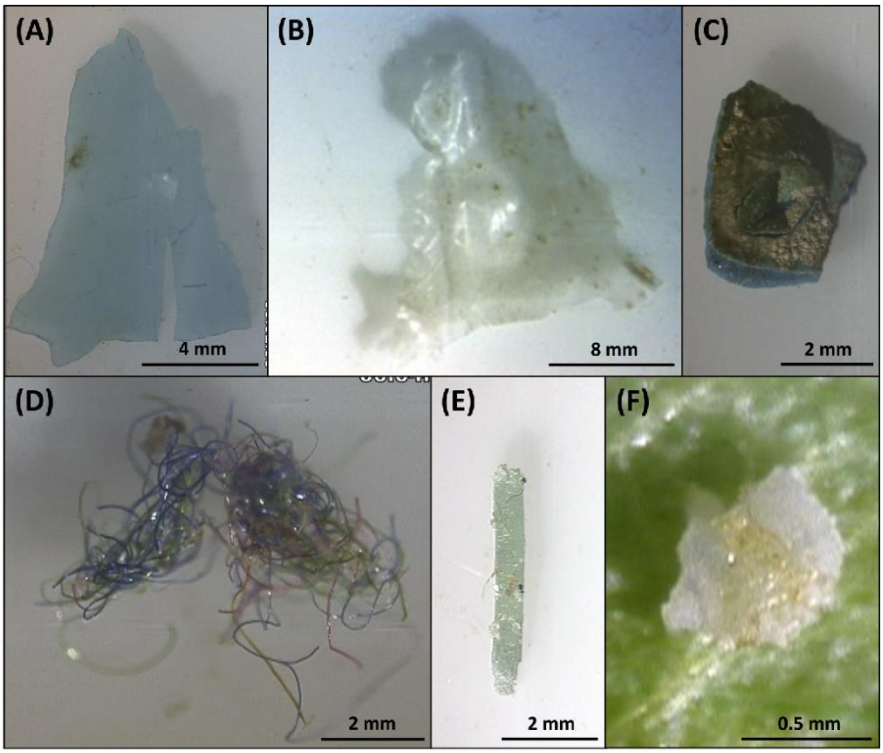

Fig. 1.4 Photographs of morphotypes of plastic debris (A) fragment, (B) film, (C) foam, (D) fiber, (E) line, and (F) pellet observed in the manta trawl samples (Free et al., 2014).

Fig. 1.5 Coastal plastic debris in Yamagata Prefecture, Japan.

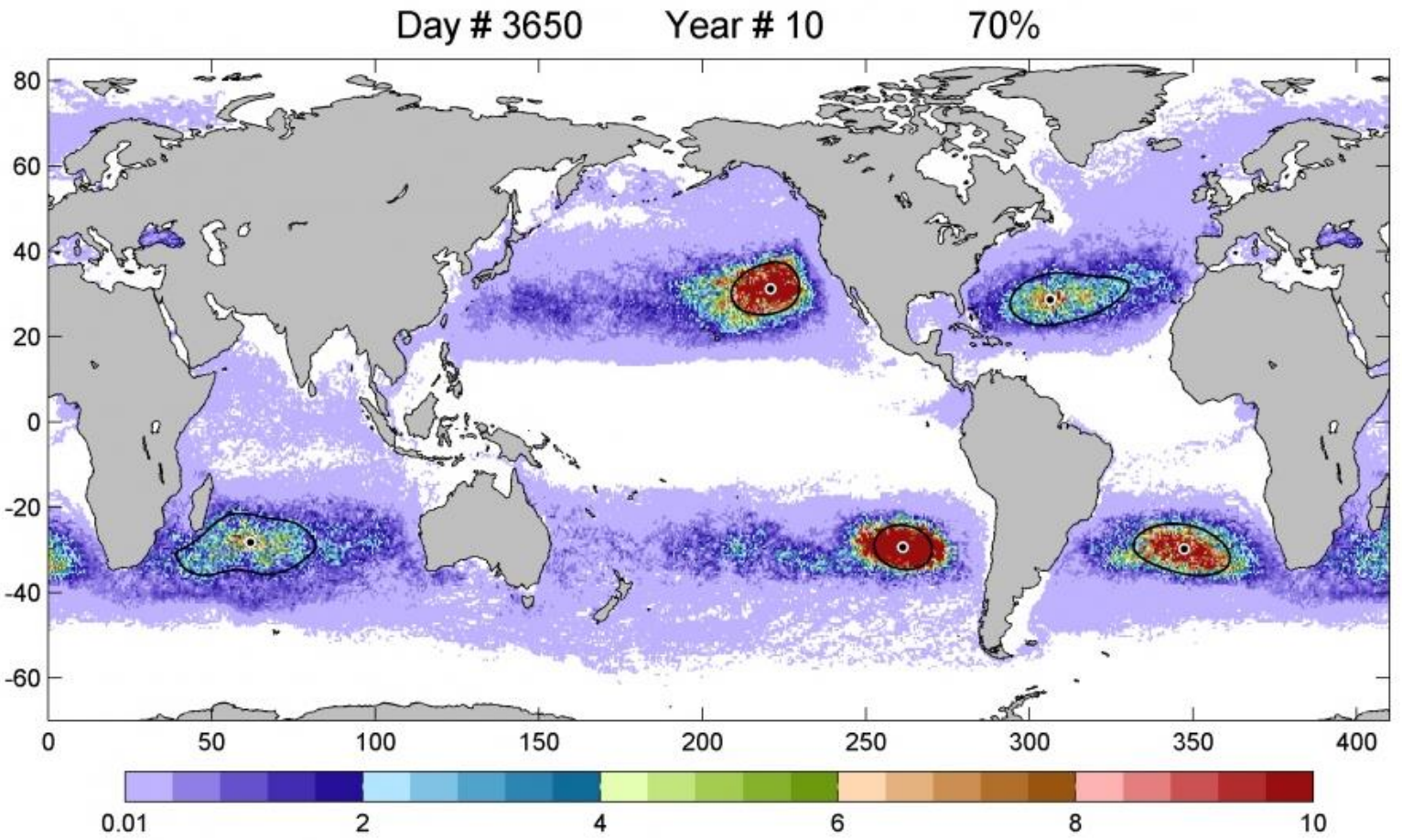

Fig. 1.6 Accumulation of plastic debris in the ocean gyres using a model based on satellite-tracked Lagrangian drifters. The model map shows the integration from the initially homogeneous state (See details in Maximenko et al., (2012)).

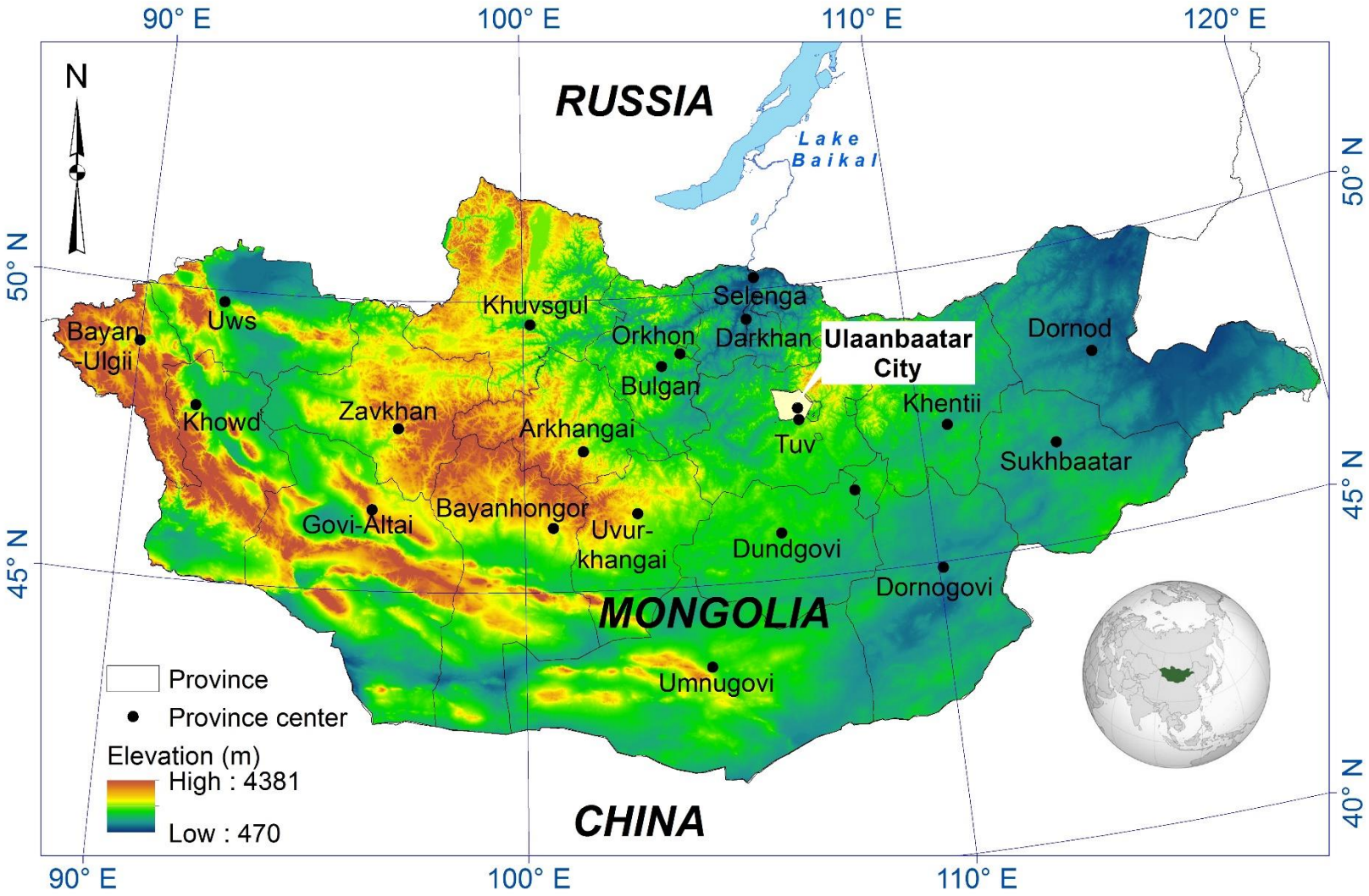

Fig. 1.7 The topographical map of Mongolia.

a.

b.

c.

Fig. 1.8 Landfill sites of Ulaanbaatar City, Mongolia (a-b) and collected waste from the public space by city sweepers (c).

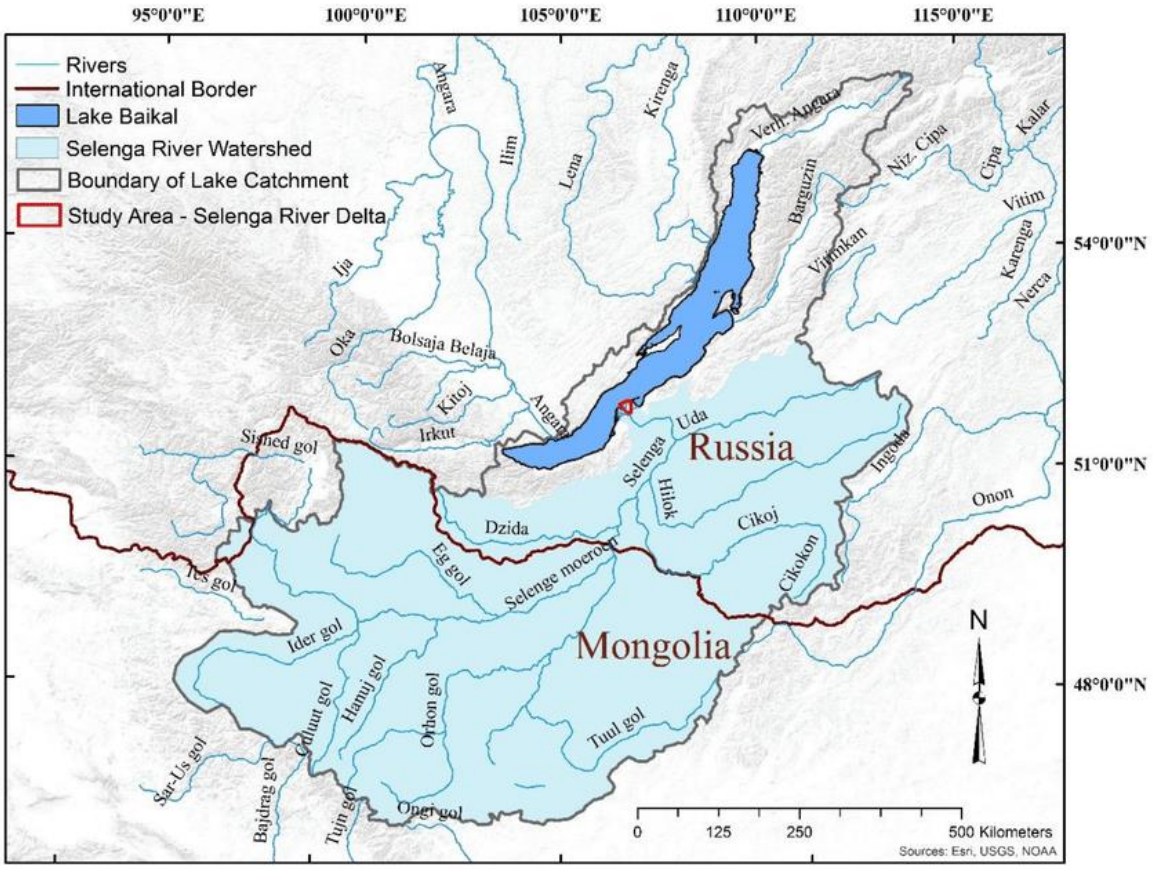

Fig. 1.9 Transboundary Selenga River basin (Berhane et al., 2018).

Table 1.1 The comparison of the average amount of plastic debris in the river environment and its predominant types and polymers in the published researches.

Target sample	Mesh size	Location	Average abundance	Dominant morphotype	Polymer type	Reference
Floating plastic debris in the top 0.5 m of the water column	500 µm	Danube River, Austria	316.8 ± 4,664.6 items/1,000 m^3	Spherules	n.d.	(Lechner et al., 2014)
Floating plastic debris in the river	300 µm	Tamar Estuary, Southwest England	204 pieces of suspected plastic (84% was confirmed)	Fragments	PE (40%), PS (25%), PP (19%)	(Sadri and Thompson, 2014)
Floating MPs in surface water	300 µm	Rhine River, Rotterdam	892,777 particles km^{-2}	Spherules (60%), Fragments (37.5%)	PS (29.7%), PP (16.9%), other types (13.6%)	(Mani et al., 2015)
Floating plastic debris in surface water	80 and 330 µm	Seine River basin, Paris	3 – 106 particles m^{-3} (80 µm) 0.28 – 0.45 particles m^{-3} (330 µm)	Fibers	n.d.	(Dris et al., 2015a)
Floating plastic debris river water	300 µm	Rhone River, Swiss	7.0 particles m^{-3} (micro) 0.012 particles m^{-3} (macro)	Fragments and foams	PE (62%), PP (15%), PS (12%)	(Faure et al., 2015)
Suspended MPs in the surface water	333 µm	Yangtze River Basin, China	4,137.3 ± 2,461.5 n/m^3	Fibers (79.1%), Granules (11.6%)	n.d.	(Zhao et al., 2014)

Mean ± SD (mean ± standard deviation); Minimum – maximum concentrations; n.d. = not determined; PE: polyethylene; PS: polystyrene; PP: polypropylene.

CHAPTER 2

Methodological challenges to explore environmental plastics

2.1 Introduction

Assessment of environmental plastics requires multiple steps including sampling, extraction, detection and quantification with different methodologies and techniques on each step (Frère et al., 2016). In the literature, various methods and techniques have been used for identification, monitoring and analyzing of plastics in the environment (Imhof et al., 2016; Li et al., 2018a). However, there are no any standardized protocols for sampling procedures and further analyses, resulting that different methods were chosen by researchers followed by difficulties of comparison between data (Qiu et al., 2016).

Analyses of environmental plastics should be usually started from sampling. During the sampling, plastics are required not only to be collected from the environment but also be separated from the environmental matrix (Rocha-Santos and Duarte, 2015). The various environmental matrices (such as water, sediment, soil, suspended solids and biota) require different sampling methods and extraction techniques (Dümichen et al., 2015).

In order to understand impacts of plastic debris, most of studies have focused on their abundance in the environment. However, as an assemblage of heterogeneous pieces of plastics with various sizes, morphotypes, shapes, colors, polymer types, specific densities, chemical compositions and other characteristics comprises environmental plastics, it is important to consider methodological criteria for quantification and identification of environmental plastic samples (Hidalgo-Ruz et al., 2012). Although a number of published literatures aim to standardize methods to obtain and determine effective separation technique, only microplastics (MPs) were exclusively targeted in the samples without any attentions of applicable protocols for the other sized plastics (Avio et al., 2017; Hanvey et al., 2017). Indeed, the pressure of MPs on aquatic ecosystems is more severe than mega-, macro-, and meso- sized plastics, however, breaking down of larger sized plastics largely produce secondary MPs with various stages of aging (Halle et al., 2017; Hanvey et al., 2017). These degradation mechanisms should be addressed for evaluation of larger sized plastics and standardized approaches for collecting and characterizing those plastics in various environmental matrices (Axelsson and van Sebille, 2017).

Since there is still no certified procedure for environmental plastics, this chapter focused on sampling, separation and determination of plastic pieces to review various methodologies for plastics in environmental systems, especially focusing to riverine environment. Additionally, this chapter summarizes and compares the analytical approaches for evaluation of plastic debris including mega-, macro-, meso- and micro-plastics in aquatic environment. Ultimately, recommendation of standardization for determination of plastic debris in the aquatic ecosystem can be offered in this chapter.

2.2 Sampling procedure

2.2.1 Sampling strategies

There are three main sampling strategies (selective, bulk and volume-reduced sampling) used for recovering plastics from the environment (Hidalgo-Ruz et al., 2012). The method of sampling is influenced by many factors, while each sampling method has its own advantages and disadvantages.

Selective sampling: Visible plastics (>1 mm in size) that are recognizable by naked eyes can be directly collected from the environment. Visible mega, macro, meso and micro-sized plastics can be often sampled by this technique, because their size range (from ~5 mm to >100 mm) allows us to find them easily in the environment. However, the major disadvantages of this technique are less obvious and heterogeneous pieces are often overlooked when particularly mixed with other debris or have no characteristic shapes (i.e., irregular, rough, angular).

Bulk sampling: An entire volume of environmental matrix (ex. water, sediment and soil) is taken without reduction of volume. Using this technique, all plastics can be sampled regardless of their sizes and visuality in the environment. Furthermore, processing bulk samples can prevent overlooking and plastic loss during the sampling. All MPs which can be easily lost and/or often covered by sediment particles can be collected by the bulk sampling.

Volume-reduced sampling: Volume of environmental matrix is usually reduced as much as possible during the sampling until reaching to a specific number or size of plastics targeted for further analyses. This technique has been broadly used for sampling through direct sieving of water and sediment on the sampling site and through filtering by passing a net for a large volume of water.

2.2.2 Plastic samples from river water and sediment

There are several different methods for sampling of plastics from the river water and sediment. The plastic sampling from water is mainly conducted by volume-reduced or bulk sampling techniques. The volume-reduced technique is mainly employed for the planned plastic debris sampling in surface water, water column and bottom water using a net or a sieve. The most commonly employed net is a plankton net (333 or 335 µm mesh sizes), although other nets with mesh sizes ranging from 50 to 3000 µm have been used in several studies in freshwater and marine environment (Hidalgo-Ruz et al., 2012). Recently, Liedermann et al., (2018) presented a new device to collect plastic samples from three different depth of large river. The advantage of this newly introduced device which installed nets with different sizes (41 µm, 250 µm, and 500 µm) is availability of vertical and horizontal samplings at the same time. However, selected different mesh sizes in the various studies provide great differences in size and abundance of obtained plastic samples from the waters and it is only available for large rivers possible to set the net (Dris et al., 2015a). The bulk sampling technique is mainly conducted for smaller sized plastic particles extracted from water using pumping or filling bottles with water. The collected water is usually passed through a filter followed by calculation for number of plastics from the fixed volume of water. In this case, mesh size of the filter is relatively lower than the nets which are used for the volume-reduced sampling technique, resulting that the captured plastic particles (micro- and nano-plastics) are several orders higher than those in the net sampling (Barrows et al., 2017). In addition, cross contamination in procedures of sampling and laboratory analysis can be minimized using a bulk sampling. However, only a few studies have used water samples collected by the bulk sampling approach (Li et al., 2018a).

Plastic samples from sediment are generally taken from river bottom and shore sediments (Klein et al., 2015; Yan et al., 2019). Based on targeted plastic debris and aim of studies, selective, bulk and volume-reduced sampling techniques can be used for the plastic sampling from the sediments. Hidalgo-Ruz et al., (2012) reviewed that sediment depths during the sampling varied from 1 cm to 32 cm and mentioned that most studies focused on top 5 cm of sediment in marine and freshwater environments. Collected sediments from the river can be sieved (1 − 5 mm sieves) on site to obtain targeted size of plastics (volume-reduced sampling) or bulk samples can be applied to the further step of plastic extraction. On the other hand, the selective sampling technique for sediment samples is usually conducted using tweezers (Ashton et al., 2010) tablespoons (Cooper and Corcoran, 2010),

or picked up by hand (Mato et al., 2001b; Van et al., 2012) for collection of plastic pellets and those fragments. Owing to its difficulty to reach, deep sediment samples were mainly collected by the bulk sampling technique using a core or grab (Kanhai et al., 2019; Turner et al., 2019), while shallow water sediments were collected by iron scoops (Corcoran et al., 2015). Bulk samples collected by grabs and scoops were also directly sent to a laboratory for any following analyses without field sieving (Castañeda et al., 2014).

Variable methodologies for sampling depending on tools and geographical backgrounds directly affect to units on data. Units are mainly based on sampled area (ex. m^2, ha, km^2), volume of samples (ex. L, m^3), and weight of samples (g, kg, t) (Hanvey et al., 2017). Given to the various unit to express the concentration of plastic in the environmental matrices, comparison between the concentrations of plastic debris in the aquatic environment is complicated. At this point, the improvement for methodological standardization could not only include sampling techniques using tools and chemicals but also international (SI) unit to express the data.

2.3 Sample preparation

Environmental matrices (water and sediment) require various preparation processes which depend on sample properties and aims of the research work. However, there are three main procedures (digestion, density separation, and filtration) for further analyses (Eerkes-Medrano et al., 2015; Rocha-Santos and Duarte, 2015).

2.3.1 Digestion and cleaning

Plastic pieces covered with biofouling (layer of microorganisms) during their exposure to aquatic environment (Crawford and Quinn, 2017). The presence of biofouling has been recognized by surface changes of plastics such as color, texture and shape. Moreover, hydrophobic accretions, biogenic residues and microbial communities can be found as adsorbed materials on the surface of plastics (Cole et al., 2011; Lobelle and Cunliffe, 2011). These adsorbed and fouled materials can be interferences to examine plastic samples, leading to misidentification under the visible and microscopic analyses and sometimes negative impacts on analytical instruments. For these reasons, cleaning of samples and digestion of organic cover is necessary to conduct during the sample preparation step.

Acidic chemical, alkaline chemical and enzymatic digestion are mainly considered for digestion and cleaning of plastic samples (Toussaint et al., 2019). The acid digestion procedure recommends an oxidative acid treatment using hydrochloric acid, nitric acid, perchloric acid and chloric acid (Enders et al., 2017; Hanvey et al., 2017). However, strong acids have influenced to the surface of plastics, resulting in modification, damage or destruction of plastics. On the contrary, alkaline chemical digestion requires usage of various alkaline solutions such as sodium hydroxide, potassium hydroxide or tetramethylammonium hydroxide (Thiele et al., 2019). Biological matrices (e.g., animal tissue and organs) have been effectively hydrolyzed by alkali digestion (Li et al., 2019a) and impact of strong base on the plastic is less than that of acid digestion. Enzymatic digestion which mainly used for samples with rich in biological materials (Toussaint et al., 2019). The biological digestion is mineralizing removal of organic matter without causing damage on plastics. Although the enzymatic solution is very safe to use, the costs for biological digestion is significantly higher than those of chemical digestion. The disadvantages are that enzymes are specific to certain types of organic matrices (proteins, carbohydrates, fats etc.) and complex organic matter may require use of several sequential steps to adequately remove organic components in the organic matrices (Toussaint et al., 2019).

The most used approach for chemical digestion of plastic debris in sediment and water samples is Wet Peroxide Oxidation recommended by National Oceanic and Atmospheric Administration (NOAA) Marine Debris Program (Masura et al., 2015). Organic matter in the samples is decomposed by concentrated hydrogen peroxide (H_2O_2, 30%) supported by iron (II) as the electron donner which is called as the Fenton Reagent (Lo et al., 2018; Rodrigues et al., 2018b; Watkins et al., 2019). After mixing the Fenton Reagent with the sample, strong reaction will be occurred by hydroxyl radicals which is strong oxidant for organic substrates (Rodrigues et al., 2018b; Tagg et al., 2017). Increasing temperature till 75°C due to the reaction will promote complete digestion of organic matter (Masura et al., 2015). Additionally, several studies apply ultra-sonication to separate plastic particles from its aggregates formed with sediment particles and to isolate adhered mineral particles from the plastic surface after the digestion (Hidalgo-Ruz et al., 2012; Zbyszewski and Corcoran, 2011).

2.3.2 Density separation

Density separation combined with filtration is the most fundamental step for plastic extraction from the sediments (Hidalgo-Ruz et al., 2012). The separation is based on differences between densities of plastic particles and mixtures in sediment. Typical densities for sand and other sediments have higher densities (approximately 2.55 g cm^{-3} and >1.44 g cm^{-3} respectively) than most of plastic materials (Hanvey et al., 2017; Hidalgo-Ruz et al., 2012). Table 2.1 shown the densities of common polymers (ex. polyethylene 0.92 − 0.96 g cm^{-3}; polypropylene 0.89 – 0.92 g cm^{-3}; polystyrene 0.28 – 1.04 g cm^{-3}; polyethylene terephthalate 1.37 − 1.45 g cm^{-3}; polyvinyl chloride 1.16 − 1.58 g cm^{-3}; etc.). Differences in densities making separation a practical technique for plastic sample extraction from the sediments. A high-density solution mixes with samples and shake it for a fixed time. After the shaking, the environmental matrix is settled for a while to leave low density plastic particles in suspension or floating on the surface of the solution (Li et al., 2018a; Rodrigues et al., 2019). The common solutions for the density separation are NaCl (max. density 2.16 g cm^{-3}), $ZnCl_2$ (max. density 2.91 g cm^{-3}), $CaCl_2$ (max. density 2.15 g cm^{-3}), NaI (max. density 3.67 g cm^{-3}), sodium polytungstate (SPT max. density 3.1 g cm^{-3}) and filtered sea water (density 1.05 g cm^{-3}) (Hanvey et al., 2017).

A solution is chosen according to the density of target plastics. The most commonly used salt solution is NaCl with the density of 1.2 g cm^{-3} due to its low cost and no toxicity to humans (Browne et al., 2011; Karlsson et al., 2017; Peng et al., 2018). However, high density plastics such as polyvinyl chloride and polyethylene terephthalate are overlooked leading to underestimation of total plastic concentration in the environmental matrix (Claessens et al., 2013). Separation solutions with various densities have been used in numerous studies to extract the plastics from the sediments. For example, Kedzierski et al., (2017) was successfully extracted MPs (93 – 98%) from sand samples in Europe using NaI solution with a density of 1.8 g cm^{-3}. However, NaI is expensive and must be handled with care (Nuelle et al., 2014). Several researchers mentioned that most of MPs recovered from the environmental matrices when using $ZnCl_2$ solution with density of 1.6 g cm^{-3} (Lahens et al., 2018; Rodrigues et al., 2018b; Zhao et al., 2014). However, $ZnCl_2$ solution is necessary to reuse and recycle in order to minimize environmental pollution by its drainage (Liebezeit and Dubaish, 2012; Rodrigues et al., 2019). Although Stolte et al., (2015) have successfully extract micro-fibers from the mineral sediments using $CaCl_2$ with density of 1.30 – 1.35 g cm^{-3} solution in the laboratory experiment, a salt residue from the $CaCl_2$ solution on the filter

makes it difficult to identify MPs on the filter. Another appropriate option is more expensive but non-toxic SPT solution with density of 1.5 g cm^{-3} (Dris et al., 2015b; Li et al., 2018a; Turner et al., 2019). All salt solutions have advantages and disadvantages to apply for plastic separation. The solution should be chosen according to efficiency, cost and safety as well as ability to use (Hanke et al., 2013). After mixing with the heavy solution, separation can be conducted under the gravity or can be accelerated by centrifugation to achieve an equilibrium for a range of density of plastic debris (Toussaint et al., 2019).

2.3.3 Filtration

The dry and wet sorting techniques (generally sieving and vacuum filtration) are the most common approaches to recover plastics from the environmental matrices (Hanvey et al., 2017). Dry sorting by sieving is mainly used to collect the plastic samples from the sediments and separate the plastic samples into targeted particle size fractions. The dry sorting technique can be applied to collect only visible sized plastics (Käppler et al., 2016). Because most of invisible plastics occurred as a solid mixture with sediment particles, density separation technique is required to separate the MP particles. The most used mesh size of a sieve is varied from 0.5 mm to 5 mm (Castañeda et al., 2014; Hidalgo-Ruz et al., 2012).

The wet sorting technique is vacuum filtration. After the density separation, floating MPs in the supernatant can be collected on the filter by vacuum filtration from the sediment matrix (Hanvey et al., 2017). Various types of materials and pore sizes of filters are applied for the vacuum filtration to recover plastic samples. The glass microfiber, cellulose nitrate/acetate and polycarbonate membrane filters with 0.2, 0.22, 0.4, 0.45, 0.7 and 0.8 μm pore sizes have been used in the reported studies (Lin et al., 2018). Although the irregular surface of glass microfiber filter (GF/F pore size: 0.7 μm) made it difficult to focus the surface of the MP particle by microscopic views, Vianello et al., (2013) succeeded to achieve a spectral analysis with good quality using a glass fiber filter. The glass microfiber filter is the most commonly applied filter to collect MPs on the filter in order to identify the polymer origin of collected environmental MPs. However, another test experiment showed that the polycarbonate membrane filter (Isopore Filter, 0.2 μm GTTP) also has a good potential for chemical imaging of MPs using FTIR measurements (Löder et al., 2015). Tested polyethylene particles demonstrated that the polycarbonate membrane filter was suitable for the Focal plane array detector-based micro-FTIR imaging. Additionally, polycarbonate

membrane filter already successfully applied on the MPs in marine sediments (Harrison et al., 2012), because of smooth and homogeneous surface of filter which makes it easy to identify MP particles and record good spectra from MP particles.

Collected bulk water samples were also filtered in this step to collect MPs on the filter instead of digestion and density separation techniques. Therefore, commonly used filters are glass microfiber and polycarbonate membrane filters. This pore size of the selected filter considered to be a limitation of size of plastic particles to recover. In addition, it is necessary to minimize sources of contamination such as airborne synthetic particles and avoid loss of plastic particles on the wall of the filter holder (Santos et al., 2015). For accurate results, both over and underestimations should be considered well during the sample preparation process.

2.4 Analysis/detection

2.4.1 Visual identification

Once plastic samples obtained from the environmental matrices, visual identification of morphological characterization has been conducted by naked eyes or under the optical microscopic view. Visual identification has been applied for visible and invisible plastic samples in order to classify size (mega, macro, meso and micro), color (black, white, translucent and colored) and morphotype (fiber, film, foam, fragment and pellet) of plastics (Andrady, 2017; Hidalgo-Ruz et al., 2012). Visual inspection can be directly applied for visible sized plastics in the research field or in the laboratory. Most studies on floating plastics carried out by visual soring of plastic samples to identify and count the collected of plastics (Sadri and Thompson, 2014; Suaria and Aliani, 2014). However, in terms of complex natural condition and heterogeneity of plastic objects, smaller MP particles (<1 mm) were difficult to distinguish plastic from other materials by naked eyes identification. For these reasons, microscopic inspection was applied for invisible plastics to prevent misidentification of particles (Vianello et al., 2018). Hanvey et al., (2017) mentioned that MPs less than 100 µm are very difficult to identify by visual identification even using a microscope. Several studies mentioned that misidentification is common for transparent and fibrous particles (Eriksen et al., 2013; Hidalgo-Ruz et al., 2012) and the frequency of misidentification increases with decreasing particle sizes. Thus, spectroscopic confirmation has been recommended for particles less than 100 µm on its longest dimension (Crawford

and Quinn, 2017). Combination of microscopy and spectroscopy (i.e. micro-FTIR) can be secure to study MP particles (Hanvey et al., 2017).

2.4.2 Instrumental analyses of plastic identification

Based on the aim of study, further instrumental analysis to characterize the environmental plastics carried out using instruments, such as FTIR, Ramen spectroscopy, scanning electron microscope equipped with an energy-dispersive X-ray (SEM/EDX) spectroscopy and pyrolysis-gas chromatography-mass spectroscopy (Py-GC/MS).

Detailed surface characterization of MP particles and elemental composition of plastics have been investigated by SEM/EDX (Coppola et al., 2016; Eriksen et al., 2013; Retama et al., 2016; Su et al., 2018). The SEM/EDX spectroscopy allows us to identify not only inorganic plastic additives such as reinforcing agent, plasticizer, coagulants containing nanoparticles of Ti, Ba, S, and Zn but also contaminants from the environment like as heavy metals accumulated on the surface of MPs (Eriksen et al., 2013; Wang et al., 2017a). Understanding of surface characteristics affected by weathering is also possible to analyze surface morphology of MPs by SEM/EDX spectroscopy (Fig. 2.1, Cooper and Corcoran, 2010; Sathish et al., 2019; Shruti et al., 2019). However, high frequency of measurements for different morphotypes of plastics, different stages of weathering and a wide range of sizes of plastics is required to understand plastic pollution of environment through researches of environmental plastics. Coupling with macro-scopic analyses will be useful to express the plastic pollution.

Recently, Py-GC/MS have been successfully introduced in plastic identification to obtain structural information about macromolecules by analyzing their thermal degradation products (Fries et al., 2013). Py-GC/MS can be utilized for the identification of MPs in the environmental matrix, as well as simultaneously identifying any present additives (Nuelle et al., 2014). Organic plastic additives that cannot be easily extracted, dissolved and hydrolyzed was identified by Py-GC/MS (Dümichen et al., 2017). The great advantage of Py-GC/MS is the direct apply of less amount of sample with minimal pre-treatment. The main disadvantages of Py-GC/MS technique are the high cost for analytical instruments. The low sample quantity (~1 mg or 100 μg) is a kind of advantage for the analysis, but concurrently one analysis is not always the representative for a heterogeneous environmental sample consisting of many synthetic polymers of different origins.

FTIR and Raman spectroscopy are the most common used methods for MPs identification (Andrady, 2017; Hanvey et al., 2017; Käppler et al., 2016). Polymers have their own fingerprint regions in the FTIR and Ramen spectra which based on specific frequency of vibration originated from chemical bonding in a polymer molecule (Käppler et al., 2016). The combination of microscopic and spectroscopic methods (micro-Raman or micro-FTIR) provide chemical structural information with high resolution. Both techniques of Raman spectroscopy and FTIR are complementary (Lenz et al., 2015).

Raman spectroscopy can measure plastic particles with very small size (below 20 μm) (Araujo et al., 2018). A monochrome laser is applied to excite an irradiated molecule, and the obtained Raman spectra provide information about molecular vibrations on the surface of the polymer (Araujo et al., 2018; Rezania et al., 2018). The applicable laser wavelength usually ranges between 500 and 800 nm. The spectra can be referred to the library to identify the polymeric status (Rezania et al., 2018). Raman spectroscopy perform better in wet samples and generate spatial chemical images of MPs (Qiu et al., 2016). Additionally, Cole et al., (2014) have successfully used the Bio-imaging techniques including coherent anti-Stokes Raman scattering microscopy to visualize the distribution of tiny MPs internalized by zooplankton. However, Raman spectroscopy does not work well with the presence of color, additives and MPs covered with contaminants (Andrady, 2017; Araujo et al., 2018).

The most common method for determination of plastics obtained from the environmental matrix is the FTIR spectroscopy due to its high spectral resolution and accurate result (Qiu et al., 2016). Using FTIR spectroscopy the plastic sample is irradiated with infrared (IR) beam (wavenumber range 400 – 4000 cm^{-1}). A part of the IR radiation is absorbed depending on the molecular structure of the sample and finally measured in transmission or reflection mode (Käppler et al., 2016; Li et al., 2018b). Plastic polymers can be identified by specific absorptions with the fingerprinting region in the obtained IR spectra. Additionally, polymer types can be quickly identified without destruction of sample (Hanvey et al., 2017).

2.5 Review of the methodologies in the river sediment and recommendation

2.5.1 Samples in the river sediment

As mentioned in the previous chapter, rivers are important pathways for plastic debris migration from terrestrial sources to marine environment, while the pathway is still unclear

because of few information of exact contribution of riverine flux of MPs (Zhang, 2017). However, it is known that plastics have accumulated for decades in freshwater systems and building up 'hotspots' of pollution (Tibbetts et al., 2018). Those hotspots polluted by plastic debris in a fluvial ecosystem can be commonly viewed as a stable recorder of current and historical contamination of plastic debris in the aquatic system. Regarding to environmental fate and behavior of the plastic debris, hydrodynamic forces (turbulence, stratification and plume fronts) influence plastic debris to follow circulation, dispersion, suspension, and settling pathways (Eerkes-Medrano et al., 2015). As well as, density of plastic fragments influences their behavior in the aquatic systems (Fazey and Ryan, 2016).

Densely populated urban environments at river mouths are great concerns as fatal areas of MPs release in the coastal areas, because river and small streams flowing through a highly populated regions should receive heavy loads of plastic pollution. In addition, it is necessary to focus plastic contamination in the river systems where locate far beyond from the river mouth to understand riverine transportation mechanisms of plastic debris from the terrestrial environment. Although floating plastics seemed to be addressed in the riverine environment to demonstrate potential transportation mechanism, it is not appropriate for sampling from the small rivers and tributaries using the volume-reduced and the bulk sampling techniques. Even collected data from the small stream samples might have large deviation which is not applicable to compare with other research results. In this case, river bottom sediments were relevant for understanding of transportation and deposition mechanisms according to the fate and behavior of plastic debris in the environment. Furthermore, 'hotspots' of pollution have been recorded by plastic debris distribution in stream networks running under changeable hydro morphological condition. For these reasons, current researches mainly focused on river bottom sediments to examine MP contamination in the high populated urban area.

Studies of MPs in river sediments are rare (Dris et al., 2015b) but a number of publications has increased during last 5 years. Unique methodologies in sampling approaches have been applied in many research works. Most of studies commonly used a grab sampler or scoops (shovels) to collect sediment samples, and analyses were carried out by microscopic and spectroscopic measurements after digestion, density separation and filtration (Horton et al., 2017a; Peng et al., 2018; Wang et al., 2017b). The sample preparation process for collected sediment samples was also conducted by a series of steps for significant recovery of plastic particles in the river sediments. Furthermore, most of studies used Wet Peroxide Oxidation method for digestion of organic materials (Ding et al., 2019; Rodrigues et al., 2018a; Van Cauwenberghe et al., 2015), indicating that contaminated

organic matter on plastic debris should be removed as plastic preparation for further instrumental analyses.

From the viewpoint of various methodologies and regarding to the aim of the current thesis, the recommended techniques for MP identification in the sediment samples are considered as follows.

Step 1: Digestion of organic matters and cleaning of plastic particles by the Wet Peroxide Oxidation method accompanied by ultra-sonication is recommended as the first step. The Wet Peroxide Oxidation method suggests controlling the heating temperature (75°C) using a temperature controlled hot plate. The digestion is necessary to continue until no organic matter in the solution. Ultra-sonication within 2 − 3 min is required for cleaning of plastic particles and separation of plastic particles from the formed aggregates with sediment particles. Heating temperature and sonication period are usually mandatory controlled during the experimental stage in order to avoid destruction of plastic particles.

Step 2: Density separation technique using a high-density solution fixed to 1.5 g cm^{-3} is required to recover heavier plastic polymer types (polyvinylchloride, polyethylene terephthalate, etc.). Additionally, mixing with the density solution and the separation process required stirring and settling time which is controlled by operation of magnetic stirring and centrifugation. For instance, the magnetic stirring for 10 min can achieve well mixing of all sediments and plastic particles. Then, centrifugation at 4000 rpm for 30 min also commonly recommended for the purpose of clear separation of all MP particles from the fine sediment particles. Finally, the supernatants are filtered through a membrane filter with 0.2 μm pore sizes to obtain high recovery of MP particles due to MPs clear imaging on the smooth surface of filter.

Step 3: Visual inspection by naked eyes and digital microscopic identification of plastics is required in this step as a qualitative and quantitative analyses of plastics. For visible sized plastics can proceed by visual inspection without microscopic identification. However, invisible MPs necessary to handle with optical microscope to characterize the physical appearance and behavior in the environment. From the visual identification, collected plastics size, color, morphotype and surface status changes such as degradation and fragmentation can be examined. During the steps 1 and 2, it is necessary to check whether collected samples are affected or not through the preparation process. To prevent cross contamination and to avoid underestimation and overestimation of plastic particles, blank samples should be evaluated for each experimental procedure in the quantitative and qualitative analyses.

Step 4: Polymer identification by a micro-FTIR spectroscopy is recommended for the final step of examination of plastic samples. In this step, the collected plastic pieces were confirmed as plastics referring to certified spectra of each polymer which could not be fully recognized in the step 3. The micro-FTIR analysis provides accurate spectral data which can be used for detailed examination of chemical structure of plastic polymer. Using the chemical structural data of measured samples, altered surface property can be identified when compared with the reference polymer data. Furthermore, altered surface property of plastics provide the surface degradation status of plastics which is one of the important behavioral information of plastic debris in the environment.

The plastic samples from these experimental stages are also able to use for further analyses tracking contamination status or changes in source of polymers during weathering processes using thermo-chemical analyses which was explained in the previous section.

2.5.2 Samples on the river shore

Most of studies on environmental plastics focus on the water, sediment and biota in marine environment, and their impacts on marine ecosystems are well documented (Thompson et al., 2009). Therefore, there are several modeling studies of plastics in marine environment and river plastic inputs to the ocean (Eriksen et al., 2014; Lebreton et al., 2018). However, approaches of plastic litter released from anthropogenic activities to rivers is not well known. Indeed, effluents from WWTPs and sewage canals were known to be a major source of MPs in the river environment. Therefore, one of the potential sources of river pollution is a large litter deposit as a result of intentionally or illegal deposition at riversides (Kiessling et al., 2019). A large-scale litter deposit center can accumulate plastic debris along the river shores by wind transport, heavy rain events, spring melt waters and deposition from the changes of river water level. The dynamics of distribution and accumulation of large plastic litter on the river shore relatively less studied as compared to floating MPs in the river environment. Furthermore, there is no specific sampling technique for plastic litter on the river shore, and no studies focused on the river shore plastics except for riverside litter at rivers in Germany (Kiessling et al., 2019). The German study performed the sampling strategy setting up three transects including three sampling circles (0 − 5 m, 5 − 15 m and >15 m from water line) in each transect to collect all visible litter on the riverside. Additionally, the study was part of an extensive project for citizen science named “Plastikpiraten (Plastic Pirates)” to investigate the litter pollution at rivers in Germany. The

sample collection of study was conducted by large number of volunteering groups under the project using selective sampling technique and all litter within the circles classified according to their types such as paper, plastic, glass, etc. (Kiessling et al., 2019).

Literally, sampling techniques for beach sediment samples can be adopted to the river shore plastics because both are technically coastal areas. Selective, volume-reduced and bulk sampling techniques are able to conduct as river shore sampling procedures for assessment of plastic contamination. However, comparing to beach sand, river shore sediment consists of various sized particles which arise the difficulties to collect volume-reduced sample through sieving. On the other hand, due to changes of hydro morphological parameters (e.g. seasonal change of water level and flow rate), bulk sediment samples from the river shore is delusion of concept, which indicate that the shore sediment is considered as a bottom sediment during a wet season with high water level. On the contrary, during a dry season with low water level, river bottom sediment near shore can be considered as river shore sediment. Eventually, the most appropriate sampling technique is considered to be a selective sampling technique to study river shore plastic pollution.

Moreover, visible plastics appear to receive particular study interests regarding to its self-degradation into invisible MPs in the coastal environments. For example, to cover all plastic debris on the beach, Lee et al., (2015) measured the macro-, meso-, and micro-plastics in the 2 cm depth sediments from the Korean beaches along high strandline using a selective, and volume-reduced sampling techniques (Fig. 2.2). The high number of micro plastics was collected from the beaches might be derived from macro- and meso-plastics through self-degradation. Taking into account invisible MPs which has not been determined in this study, the process of self-degradation can be evaluated from the relationship among size fractions, especially between visible and invisible micro plastics. The investigation of visible sized plastics relatively less targeted in the riverine environment (Kiessling et al., 2019). Giving the respect to the invisible MPs in the river environment, it is necessary to investigate concurrently visible sized plastics on the river shore in order to examine and understand the level of plastic pollution in riverine ecosystem condition.

The movement of plastics along the shore will greatly impact their distribution and accumulation at locations along the shore. Recently, many studies tend to concentrate MPs in river sediment (Hanvey et al., 2017; Van Cauwenberghe et al., 2015). However, without quantifying the debris along the river shore, a true evaluation of the plastic abundance cannot be achieved in river plastic loads. To achieve the goal of comprehensive understanding of behavior of plastic debris in the terrestrial river environment, current thesis covered the

plastic samples from river shores and river sediments. Furthermore, collected plastic samples were identified their properties using visual and instrumental methods following the previously recommended steps (Subsection 2.5.1).

Furthermore, sampling period is also addressed from the viewpoint of hydrological background of the river. Change in rain fall and water discharges affecting to the hydrological condition could influence the sampling method which also affects unit of data. Based on differences between hydrological parameters and plastic transportation by river, several researches focused on plastic distribution before and after the flooding season (Phuong et al., 2018; Stolte et al., 2015) in order to collect representative data to express river plastic flux. In the Selenga River basin, the precipitation relatively high in summer season (Fig. 2.3). To collect distributed plastics on the river shores and river sediment, the sampling has been addressed on August and next March for further evaluation of plastic accumulation and distribution at the Selenga River system. Based on precipitation and water discharge, March could be reasonable sampling period for the settled plastics on the river shores and sediments because of the lowest water discharge accompanied by the lowest water level (Fig. 2.4). August is usually the season following the highest discharge recoded on July, resulting that floating plastics on the river surface can be left on the shore (Fig. 2.4). The behavior of plastic debris along the river side can be observed on this season. Especially the year when the research conducted was relatively dry and drought in this decade from the regional weather information. For this reason, the summer sampling is considered as a reasonable sampling period for plastic sample collection and representative samples on the river shores to express behavior and distribution of plastic debris in the river environment.

In this chapter, analytical approach for measuring plastic debris in aquatic environment have been summarized. From the viewpoint of plastic samples from sediments and river shores, particular four-step of determination technique has been recommended. Regarding to geographical background and hydrological changes, potential sampling and sample processing is subjected to challenge for the further method development and harmonization in the field of aquatic plastic pollution. Finally, method validation should be conduct using a laboratory blanks in order to improve quality and accuracy of research results.

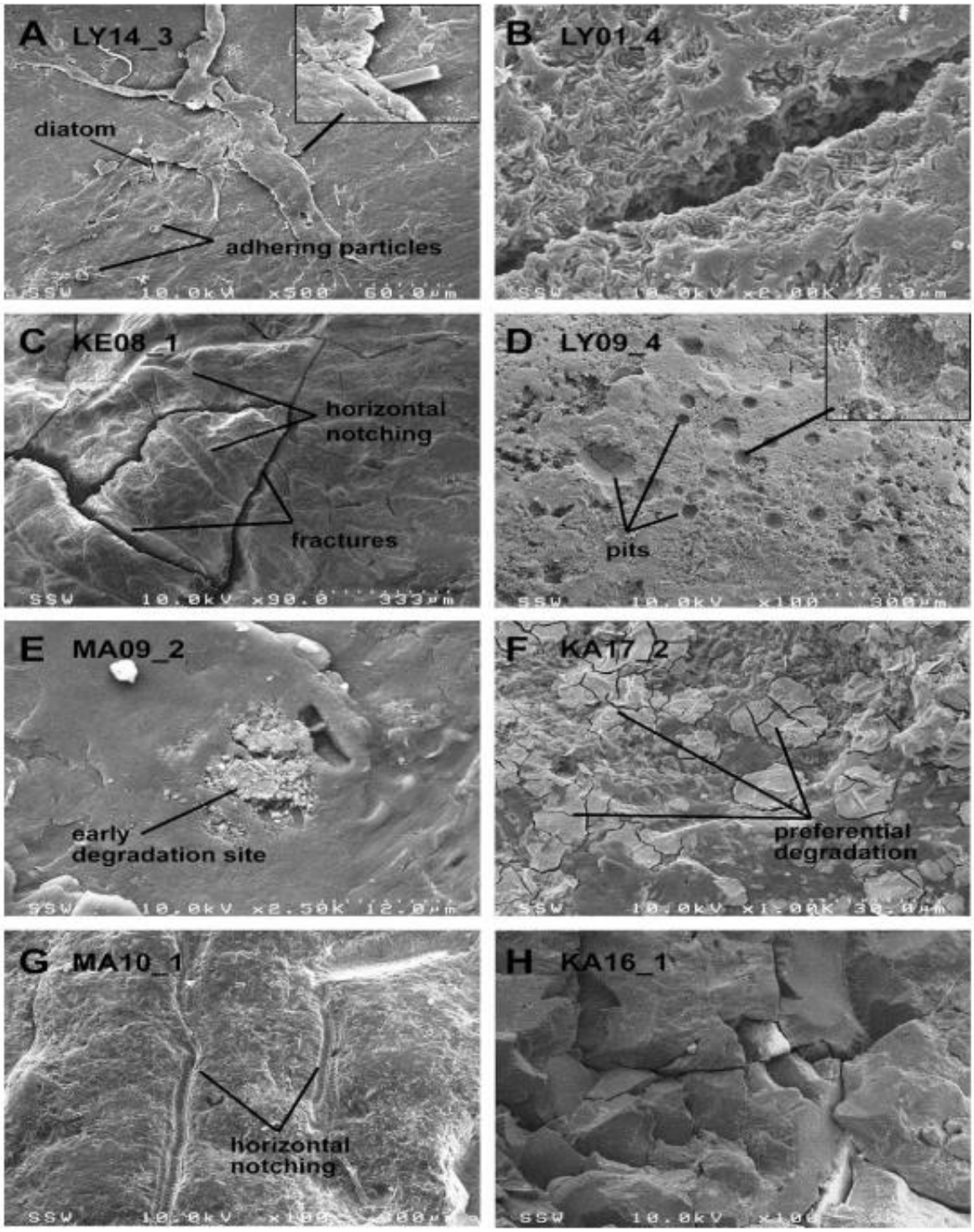

Fig. 2.1 Scanning electron microscope (SEM) images of surface textures on plastic samples. Surface changes of plastic items such as surface crack, surface degradation, notching, fractures, cracking and pits were clearly seen by SEM images. Descriptions using capital letters with number are sample ID of the photograph (See more details in Cooper and Corcoran, (2010)).

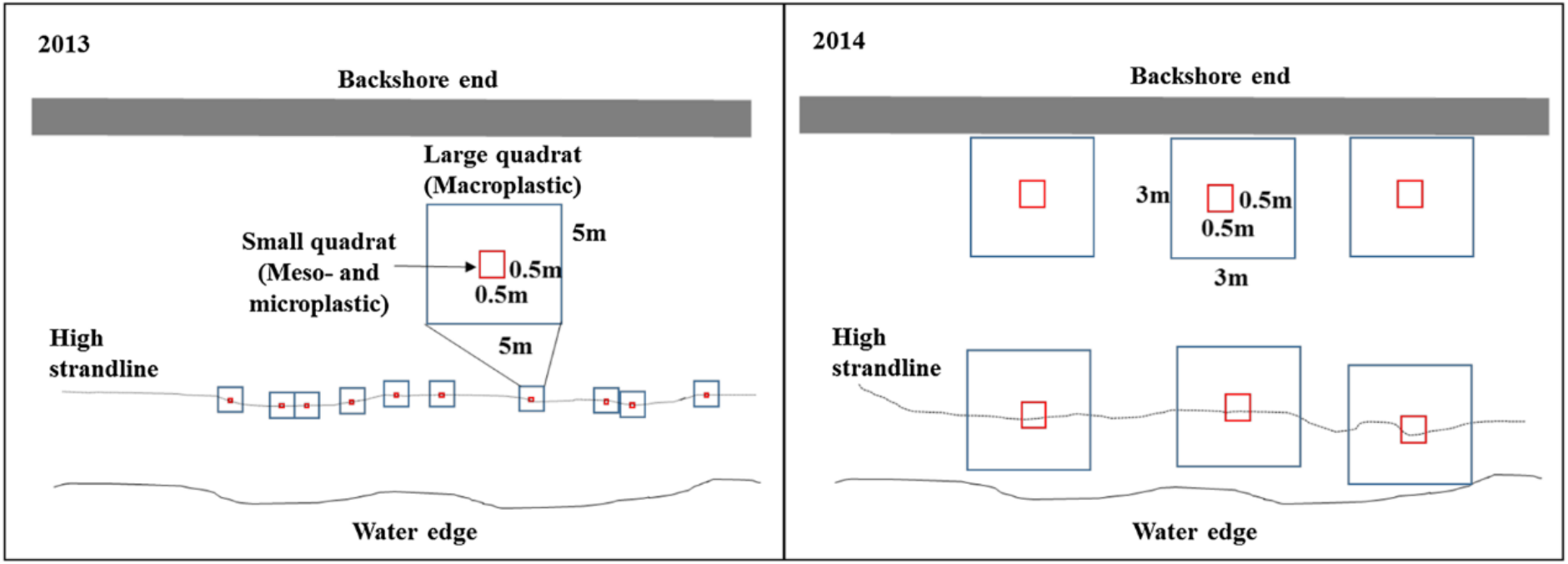

Fig. 2.2 An example of transect sampling of plastic survey on the beach from high strandline (Lee et al., 2015).

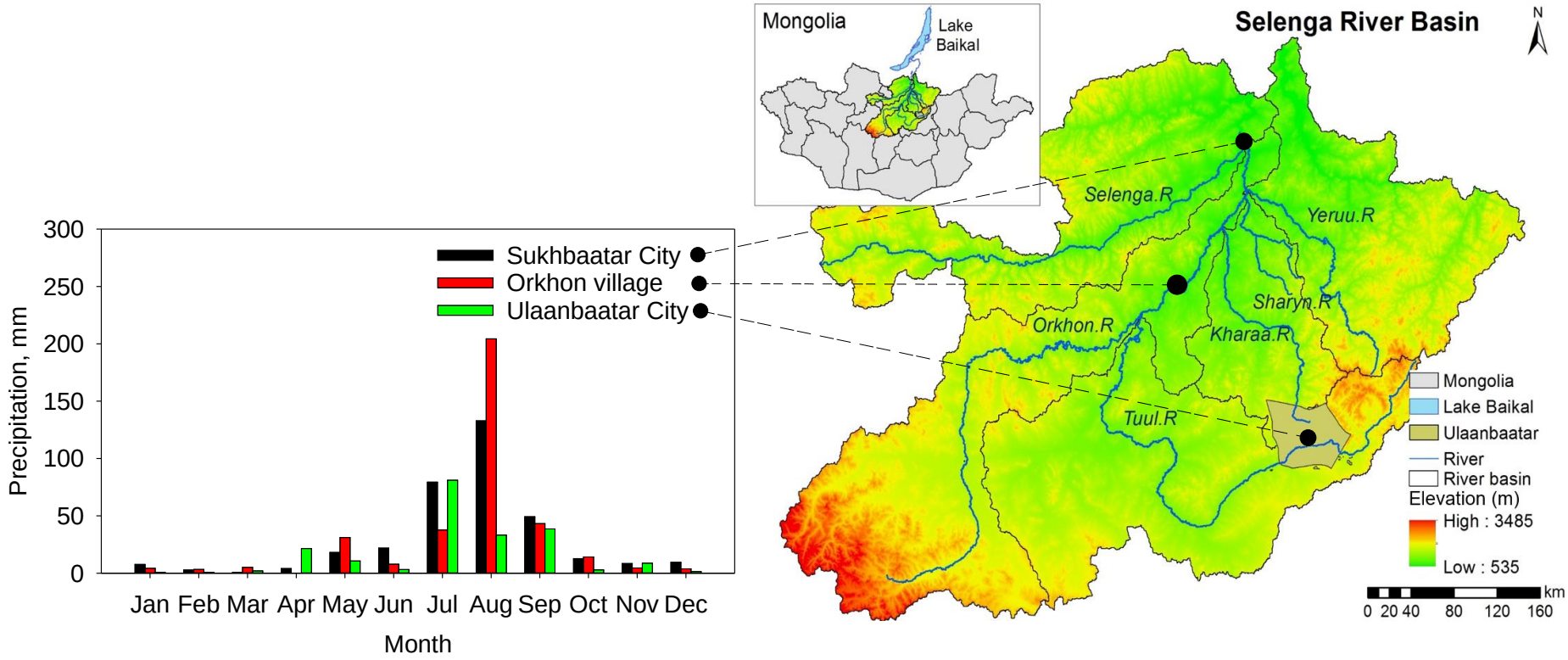

Fig 2.3 A monthly average precipitation of Selenga River basin at the observatory in the Ulaanbaatar City, Orkhon village and Sukhbaatar City with basin map (Statistical data from MongolianStatisticalYearbook-2015).

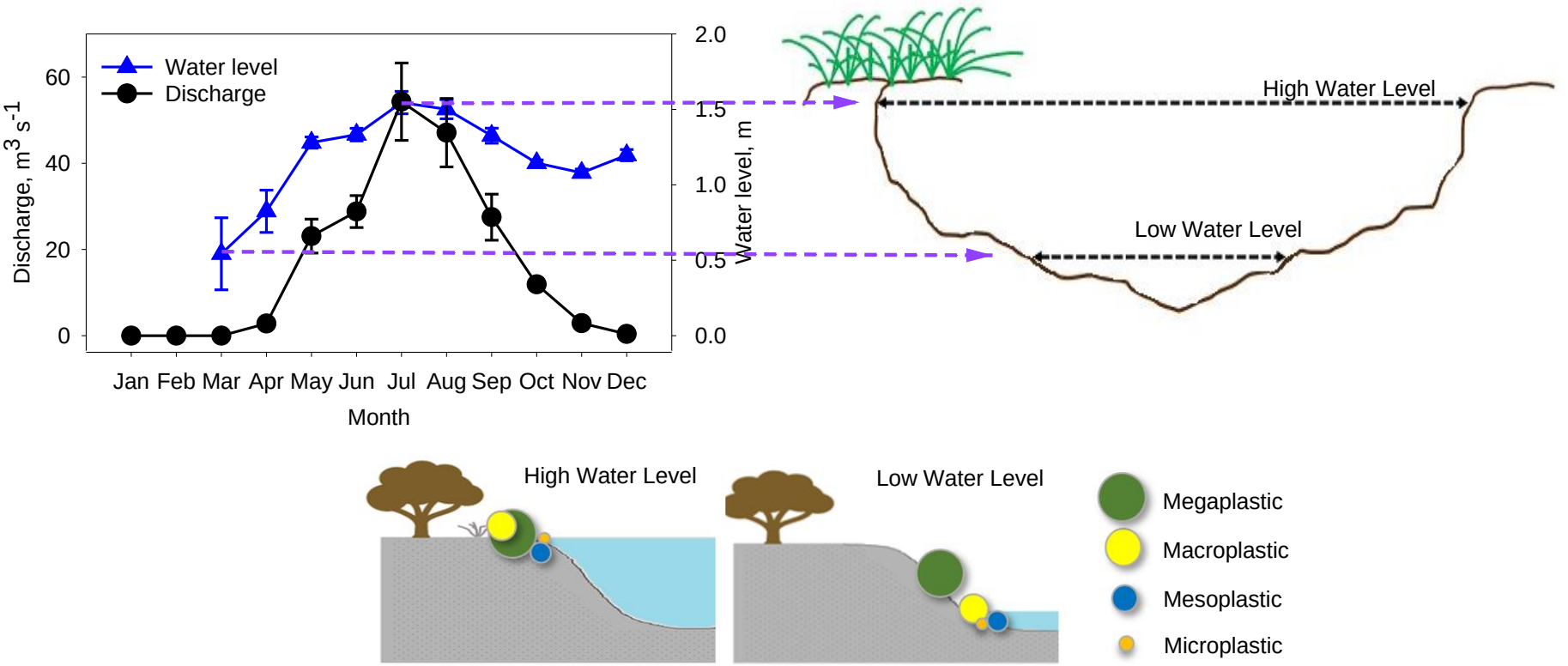

Fig. 2.4 The 5-year average river discharge and water level of the Tuul River at the Ulaanbaatar hydrological station (2011-2015) (Statistical data from MongolianStatisticalYearbook-2015). Error bar indicates standard error. The illustrations indicate differences between water level and conceptual plastic accumulation on the shore and on the bottom.

Table 2.1 Densities of polymers (Hidalgo-Ruz et al., 2012).

Polymer type	Polymer density (g cm^{-3})
Polyethylene	0.92 − 0.96
Polypropylene	0.89 – 0.92
Polystyrene	0.28 – 1.04
Polyamide (nylon)	1.02 − 1.05
Polyester	1.24 − 2.30
Acryl	1.09 − 1.20
Polyoxymethylene	1.41 − 1.61
Polyvinyl alcohol	1.19 − 1.31
Polyvinyl chloride	1.16 − 1.58
Poly methyl acrylate	1.17 − 1.20
Polyethylene terephthalate	1.37 − 1.45

CHAPTER 3

Spatial distribution and behavior of plastic debris in Mongolian river system

3.1 Introduction

Owing to plastic accumulation along coastal shores, plastic debris on the beach has been considered as the principal source of floating debris in the oceanic environment (Frias et al., 2010). However, plastic debris on the river shore has barely known in terms of environmental plastic pollution.

Most of river plastic studies mainly focused on floating microplastic (MP) debris. In some cases, the number of items that have already moved within water were also quantified (Kataoka et al., 2019; Lechner et al., 2014). As a result of seasonal events such as high rainfall and subsequent flooding, strong wind, the deposited litter in the riversides can be mobilized (Kiessling et al., 2019) and transported by river flow which is one of the major sources of floating plastic debris in the riverine ecosystem from the surrounding environment. With the exception of environmental factors, seasonal anthropogenic activities such as summer camping and winter recreation along the river, increase the accumulation of plastic litter on the river shores through unintended waste release by human activities with less control of waste disposal, and affect to the floating debris during the flooding and snow melting seasons. Those seasonal influences on the abundance of plastic litter in the river environment have been highlighted in several studies (McCormick and Hoellein, 2016). Regarding the large amount of plastic transportation by river into open water system, it is necessary to evaluate the abundance of plastic debris along the river shores in the terrestrial-aquatic system.

In recent years, the number of publications about spatial distribution of plastic debris in the river system has increased especially concerning to the plastic contamination, the amount of land-based plastic input to the open water system and their effects in the aquatic ecosystem. Although the migration of plastic debris in the inland river systems has relatively less studied compared to coastal aquatic environments. Recent studies demonstrate that behavior and origin of floating plastic items have been affected by poorly managed municipal plastic solid waste in the environment. Since various environmental and anthropogenic factors influence

the accumulation of plastics in the environment, current study targeted to evaluate abundance and behavior of plastic debris on the shores of the Selenga River system.

In order to investigate the behavior of plastic debris on the river shores of Selenga River system, current chapter aimed to ascertain the spatial distribution of visible sized plastic debris along the river and evaluate the fragmentation of plastic debris on the river shores. Furthermore, survey of plastic debris on the river shore has proposed to investigate the plastic accumulation thorough a year. This chapter emphasizes the importance of rivers as the carriage system of plastics and the distribution pattern of plastic debris along the river shores.

3.2 Materials and methods

3.2.1 Study area and sample collection

The study area is located in the Selenga River basin which lies from the central to northern part of Mongolia (Fig. 3.1). The total catchment of Selenga River basin is approximately 447,000 km^2 area including transboundary waterway from the Mongolian territory to Russian territory (Nadmitov et al., 2015). The predominant part (roughly 63%) of Selenga River basin is located in Mongolia which occupies around 282,349 km^2 area. Major rivers of central and northern part of Mongolia, contribute to the Selenga River and comprise the largest river system in Mongolia. The semi-arid continental climatic features are the typical climatic condition in this area with the –28 to 45°C of annual temperatures. The average annual precipitation in Selenga River basin is ranging from the 250 mm to 330 mm. The most of precipitation occurs between May and September. Northerly to northwesterly winds are dominant in this region (Guttikunda et al., 2013).

Since floating plastic debris carried on the highest flow season have been left on the shore with decreasing water level on dry and/or drought season, the sampling methodology was set on the river shore to examine the plastic debris in the river environment. Study shores of rivers were covered by various kind of feather grass vegetation and broader particle sizes of stones. The standard methods such as NOAA (Masura et al., 2015) and Group of Experts on The Scientific Aspects of Marine Environmental Protection (GESAMP, 2015) for plastic distribution density at large rivers and sandy seashores were not applicable in this study due to differences of river geographical background. As pointed out in the Chapter 2, the only

way to evaluate plastic distribution on river shores was to collect all visible plastics in a unit area.

Twelve sampling sites were selected from the sub-basins of Selenga River system to investigate the abundance and spatial distribution of plastic debris on the river shores (Fig. 3.1). Plastic samples were taken from the shores of Orkhon, Kharaa, Yeruu, Sharyn, and Tuul Rivers where ranging between the 47 − 50° N and 103 − 107° E from the capital city Ulaanbaatar to Sukhbaatar close to the border between Mongolia and Russia in August 2017. Sampling sites were selected based on differences of geographical background to comprehend the relationship between plastic abundance and population density in the river catchment. Owing to more than half of Mongolian population and the main industrial districts concentrated in this river catchment, selected study sites can be representative research spots for distribution pattern of plastics and their behavior in the river environment.

The sampling sites T1 to T3 are located at the upper stream in the river basin and included in Ulaanbaatar City. The shores were occupied by large stones, and feather grass vegetation was sparsely distributed on fine earth ground. The site T4 is at the conjunction of the two tributaries Tuul and Orkhon in the village Orkhontuul. The shore is muddy with low frequency of stones and is covered with less vegetation. Five sampling points (O1 to O5) were set along the Orkhon River. The sites O1, O2, and O3 are located at conjunctions with Kharaa, Sharyn and Yeruu rivers, respectively. The shores of O1, O2 and O3 are stony and covered with less vegetation. Both O4 and O5 sampling sites are in Sukhbaatar City, which is near the border between Mongolia and Russia. The shores of two sites are sandy with low frequency of stones. Feather grass vegetation was sparsely covered on the sampling quadrates. The three sites Ye, Sh, and Kh are located at the lower tributaries of the Yeruu, Sharyn, and Kharaa rivers, respectively. The shore of Ye is occupied by boulders with grass vegetation. The Sh and Kh sites were sandy shores covered with sparse grass vegetation.

Visible plastic debris was collected from the river shores within the replicated 100 m^2 area. An example of river shore was shown in the Figure 3.2 with the collected plastic samples in the 100 m^2 area. Hand-sorting technique was conducted for the plastic sample collection from the river shores and complete collection of plastics were carried out by trained persons who familiar with MPs. Triplicate measurements enabled us to calculate the average number of plastic pieces and standard deviations for each sampling site.

3.2.2 Sample sorting and identification of plastics

The collected plastic samples were counted and classified based on their size and morphotypes in the field. The size of the collected plastics was classified into mega- (>100 mm), macro- (>20 mm), meso- (5 – 20 mm), and micro- (<5 mm) size fractions (Suaria and Aliani 2014; Lee et al. 2015). Furthermore, the plastics were classified into four morphotypes, such as fiber, film, foam, and fragment (Davis and Murphy, 2015; Horton et al., 2017a), based on their appearance and characteristics. The plastic polymer types including polyethylene, polystyrene, polypropylene, high-density polyethylene, polyethylene terephthalate, polyurethane, synthetic rubber, nylon and other polymer types were categorized and identified in the collected samples in order to understand the origin of plastics. Population density data for corresponding cities, villages and administrative districts, and other statistical information were collected from the National Statistical Information Center in Mongolia (National Statistical Office of Mongolia 2015).

3.2.3 Data analysis and statistics

Principal component analysis (PCA) and hierarchical cluster analysis (HCA) were conducted using IBM SPSS 22 (IBM Co Ltd.). The Ward's method was applied to extract principal components from collected data for PCA to characterize the distribution of plastics in the research area. The HCA was used to categorize the research sites based on the distribution and composition of plastic debris. Correlation analysis was carried out using Microsoft Excel to identify statistically significant relationships between the sizes and morphotypes of the plastics.

3.3 Results and discussion

3.3.1 Total abundance and spatial distribution of plastic debris

The collected plastic debris on the river shore ranges from 2 to 506 pieces per $100m^2$ area with an average number of 134 pieces/$100m^2$ area in the study area. The average number of plastic debris and the dominant polymer type in each size fraction was shown in the Table 3.1. Micro-sized plastics recorded a relatively large number reaching to 120 pieces/$100m^2$ area on average. Fragmentation of larger plastics, which is one of the important sources of

MPs in the environment (Auta et al., 2017), might be a significant reason of widespread MPs in this river system. Predominantly occurred polyethylene and polystyrene types of plastics in this river shores (Table 3.1) are one of the widely produced and consumed polymers in the world (PlasticsEurope, 2016). In spite of size distribution, the polystyrene plastics which occupied 71.1% of identified polymers is mainly identified as foamed material in the river shores, whereas the polyethylene which occupied 16.4% of identified polymers is commonly found as plastic bags in the study area. Besides these major polymer materials, various polymer types were detected in the river environment. The detected plastic materials were polypropylene, polyethylene terephthalate, high-density polyethylene, polyurethane, nylon, synthetic rubber and other types of plastics, and they occupied 2.9, 0.9, 0.1, 0.9, 4.3, 0.2 and 3.2% to the total plastic debris, respectively.

Previous studies also frequently reported films and foams as dominant types of plastic materials in aquatic ecosystems. Lee et al. (2015) found an average number of 919.1 particles m^{-2} consisting of micro- (880.4), meso- (37.7), and macro- (1.0) plastics per m^2 at high strandlines of beaches in South Korea. The composition of plastic debris in Korean beaches indicated the dominance of styrofoam and fibers in micro- and meso-sized, and macro-sized fractions. Blettler et al. (2017) recorded the 729.97 items m^{-2} plastics consisting of micro- (704), meso- (25.1), and macro- (0.87) items per m^2 area with the dominance of hard plastics, foams and films in the shoreline sediment of Setúbal Lake in South America. Both studies on South Korea and South America explained that the film types of plastics were originated from the polyethylene polymers, whereas broadly used aquaculture, floating and insulator foamed materials that found in aquatic ecosystem were derived from the polystyrene polymer material.

Figure 3.3 showed the distributions of size and morphotype of plastics at each research site. An extremely high number of plastics were observed at the T1-T3 sites located in the Ulaanbaatar City. Additionally, sampling sites close to cities of Darkhan (Kh) and Sukhbaatar (O4 and O5) recorded the relatively high number of plastics. However, sampling sites (T4, O1, O2, O3, O4, and Sh) located at remote locations from the populated areas recorded small numbers of plastics. Mega- and macro- size plastics are common at sites with low number of plastic debris. On the contrary, meso- and micro- size plastic debris were the typical sizes of plastics at sites with a high number of plastics. The decreasing trend in the rate of MPs was observed from the urbanized areas to the low populated areas. The large number of MPs in urban areas indicate that there are numerous sources of plastic debris in highly populated areas and those plastics have been broken down into smaller size fractions

on the river shore. The source and distribution of plastics could not be clearly identified in the regions with lower population; however, plastics were still distributed from several pieces within a 100m^2 area, which were probably carried by river currents and wind from urbanized areas. In the marine environment, researches have suggested that the accumulation and distribution of plastic debris are affected by climatic forcing and other environmental effects relating to ocean (Browne et al., 2010; Kuboto, 1994; Martinez et al., 2009). In coastal areas, movement of water caused by wind, waves, and tides transport the particles of sediment according to their size, shape and density (Le Roux, 2005). Owing to various sizes and shapes of plastic debris, similar processes occurring in sea sediments which are controlled by weather condition and river hydro-morphology can control their behavior in the terrestrial aquatic system. For example, wind-driven movements considered to be significant to transportation of mega- and macro-plastic debris confirmed in this study because the larger volume plastics are easily blown away from the origin compared to the smaller volume plastics such as meso- and micro-plastics which are hidden under the vegetation.

Films, foams, fibers and fragments were identified in the study area with various abundances (Fig. 3.3b). Resin pellets represent common plastic debris in coastal and oceanic environments (Antunes et al., 2013; Mato et al., 2001a), but they were not found on the studied river shore because of lack of plastic processing factories in Ulaanbaatar City. The predominant type of plastic from the average distribution of the morphotypes (Fig. 3.3) was foams (75.6%) followed by films (14.7%), fragments (8.0%) and fibers (1.7%). The composition of plastics can be attributed to high production and consumption with respect to the population density (Fig. 3.4), and anthropogenic activities and land uses in the studied river catchment. Furthermore, the regional environmental background including climatic condition (i.e., relatively cold condition in winter) is a possible reason for high abundance of polystyrene foam (PSF) originated from foam plastics used for heat insulator in the study area. Large number of foam plastics mainly occurred in the micro size fraction in the studied river shores, differing from the results indicated by predomination of resin pellets in coastal areas (Ashton et al., 2010; Karkanorachaki et al., 2018; Le et al., 2016).

Geographically, most of landfills in Mongolia commonly locate at upland areas of mountain hills on the south facing slope in the river catchment as mentioned in the Chapter 1. Similarly, villages and populated areas were also mainly located on the south facing slope of the river. Based on downwind location, the lowland of river basins is the main receiver of plastic debris by wind transport. On the other hand, direct input of waste plastics has

recognized in the river basin through littering due to improper waste management and inappropriate human behavior. Accumulated plastic debris in the river watershed again can be washed into the river and transported with river water during the snow melting, rainy and flooding seasons. However, accumulated plastic debris can be left on the same location and break down into smaller pieces under the freeze-thaw and dry-wet cycles in winter or dry seasons. Additionally, plastic properties such as size and specific density of polymer influence on distribution, degradation status and fragmentation of plastics in the environment (Cheung et al., 2016; Hoffman and Hittinger, 2017). Given to the various influences on the behavior of plastic debris, it is important to highlight both anthropogenic and environmental factors affecting on abundances and spatial distribution of plastics and can be considered as a driving and controlling factors for the behavior of plastic debris in the river system.

3.3.2 The relationship between abundance of plastics and population density

Figure 3.4 shows a significant correlation between the plastic abundance and population density ($R^2 = 0.949$). It is clearly shown that total number of plastics is always higher on the river shores close to the city center such as sampling sites of T1, T2, and T3, Kh and O5, while the river shores in the low populated areas along the Orkhon River recorded low number of plastics. There are few larger residential areas along the Orkhon River, resulting in a small number of released plastics along the river shore. Populated areas are the considerable sources of plastics along the river shores. Browne et al. (2011) and Andrady (2017) have reported the significant relationship between number of plastic debris and population density. Furthermore, Wang et al. (2017b) pointed out that the abundance of MPs in freshwaters in China is directly related to the distance from the urban center ($r = –0.895$, $p < 0.001$).

Lakes and coastal regions were mainly highlighted by relationships between urbanization and plastic debris in the open water system (Eriksen et al., 2013; Free et al., 2014). In addition, estuaries of coastal rivers have been targeted by most of studies for riverine plastics because of heavy load of plastics from the large cities such as Los Angles, Tokyo, Chicago and New York which lay on the coastal areas, and direct input of plastic debris into the oceans. For example, inflow processes of MPs in the river environments have been evaluated in 29 rivers which were located on the coastal areas of Japan (Kataoka et al., 2019). A study of urban rivers in the Chicago metropolitan area has demonstrated the abundance of MPs in surface water and their potential to retrain and move to the downstream

habitats (McCormick et al., 2016). The concentrations of plastic and non-plastic microfibers have been evaluated in the Hudson River located in New York State of USA (Miller et al., 2017). Furthermore, the abundance of MP and the impact of municipal WWTP effluents on the MP concentration in Raritan River located in central New Jersey, US, have been evaluated by Estahbanati and Fahrenfeld, (2016). However, plastic debris in inland river systems has started to receive attention regarding to the fate and dynamic distribution of plastic debris in the terrestrial and aquatic environment. Owing to the comprehensive attention, studies demonstrate that behavior of plastic items in the inland river system has been significantly affected by surrounding environment mainly characterized by population density in the river catchment (Lahens et al., 2018; Shruti et al., 2019). Further transport of plastics might be controlled by climatic factors and the transportation process mainly is characterized by the distance from the urban district. Although there is no significant relationship between the distance from the urban district to plastic waste deposits, this study also indicates a larger number of plastic wastes on the river shore close to urban districts than that observed at any remote research fields.

3.3.3 Fragmentation of plastic debris in the river environment

Significant positive correlations were observed among average numbers of micro-, meso- and macro- sized plastics in this river system (Fig. 3.5 and Table 3.2). Fragmentation of larger plastics to smaller ones contribute the further trends in the relationships of plastic abundances between visible and invisible plastics (Barnes et al., 2009). Despite of plastic durability in the environment, plastic pieces on the river shore break down into smaller sizes by aging through freeze–thaw cycles, and physical abrasion by wind and compaction processes (Andrady, 2017, 2011). This is particular evident on aquatic shores where photo-degradation, weathering and abrasion through wave actions and water currents make plastic items brittle, increasing their fragmentation (Fok and Cheung, 2015).

In addition, the foam and film types of plastics demonstrated significant linear correlations between the size fractions (Table 3.2). The relationships between size fractions indicate that on-site fragmentation of plastics leads to a consistent size composition on the shore. Figure 3.6 showed the graphical illustration of fragmentation of plastic materials with an example of photographs of macro-, meso- and micro-sized PSFs'. From the results of significant correlations between the size fractions of foams and films, these plastics were relatively fragile against the environmental forces compared to other two types of plastics as

fragments and fibers. Andrady (2011) and Mattsson et al. (2015) mentioned that plastic degradation processes such as photodegradation, thermal degradation, and mechanical/physical degradation have enhanced the potential of fragmentation of plastics in the environment. These degradation processes were mainly driven by environmental factors such as solar radiation, temperature changes, river hydrodynamic conditions (Cooper and Corcoran, 2010; Gewert et al., 2015; Weinstein et al., 2016).

To understand the relationship between various sizes of films and foams and population density, correlation analyses size fractions were conducted with the population density of cities, villages and administrative districts (Table 3.2). Statistically significant correlations ($p < 0.001$) were observed between the size fractions of film materials (Table 3.2a). Meso- and micro sized films showed the significant correlation coefficient with the population density ($p < 0.05$), indicating that the film type plastics were fragmented from the larger-size to smaller sizes and accumulated on-site at the high population areas. In addition, micro-films originated from other three larger size films were probably broken down to invisible smaller-sized plastics and transported by river water to far locations from urban areas. This phenomenon smears the relationship because of the lower correlation coefficient compared to meso-sized films with population density. The reason for the low content of mega- and macro-films is that those large-sized films on the river shores near the boundary were cleaned by volunteers or city sweepers. On the other hand, stronger correlations with significant correlation coefficients were observed between the size fractions of foamed plastic materials ($p < 0.01$; Table 3.2b). Furthermore, the population density is significantly correlated with macro- ($p < 0.01$), meso- ($p < 0.01$), and micro- ($p < 0.001$) sized foams. On-site degradation of PSF can occur on the river shores because of brittleness of aged PSF.

In contrast, no significant relationships were observed for the size fractions of another two types of plastics (fragments and fibers) probably due to their strong physical properties compared to the films and foams. However, in this study, there are no significant relationship between visible plastics in upstream and in downstream study fields, indicating that fragmentation of larger plastics can occur on-site under natural conditions due to various physical and mechanical forces.

3.3.4 Characterization of plastic distribution

The PCA was utilized to simplify the variables and evaluate patterns of plastic distribution in the study area probably relating to their behavior in the environment using

obtained data coupling with social statistical data. Especially, PCA method is mainly used in this study to promote detailed understanding of plastic behavior and their different pathways in the environment. In this study, parameters of plastics properties (size and morphotype) and population density was evaluated by PCA analysis. Two principal components were extracted from the PCA (Table 3.3, Fig. 3.7a) with 94.4% of the eigenvalue (Ev = 7.6). As shown in component matrix (Table 3.3), the first component (63.2%, Ev = 5.1) is plastic debris in urban districts, which strongly depends on macro- (0.897), meso- (0.984), and micro- (0.990) sized plastics; foam type (0.993); and population density (0.973). The PSFs from the construction or manufacturing processes and home garbage used as packaging materials are the major sources of this urban distribution of plastics. The three size fractions in the first principal component are predictable for a fragmentation process during transportation along the river.

The second component (31.2%, Ev = 2.5) is characterized by mega-sized plastics (0.943), films (0.725), and fibers (0.958; Table 3.3). This component described the plastic debris associated with remote locations because of the low contribution of population density. Large plastic debris including mega-films and mega-fibers (i.e., plastic bags, sandbags, and plastic ropes) are the main component of distributed plastic debris. Many illegal dumping along gullies on hills in the Ulaanbaatar City found during the field survey can promote distribution of plastic debris. The number of mega- and macro-sized films (such as sheets and plastic bags) is higher than that of meso- and micro-sized films in scarce population regions, indicating that the mega-film litter in and around urbanized areas is aerodynamically transported from the upstream through the river valley by water flow and wind (Barnes et al., 2009). Browne et al., (2010) also demonstrated a similar of wind-blown plastics in the estuarine shorelines of Tamar estuary in UK. For macroplastics, distribution patterns can be clearly explained by downwind. The research report also mentioned that less density of macroplastics were the common debris distribution in downwind areas, whereas the distribution of smaller MPs demonstrated the clearest pattern of plastics with higher density (e.g., polyvinyl chloride) (Browne et al., 2010).

Based on the two components, the plastic debris along tributaries concentrated in urbanized areas due to anthropogenic activities such as improper waste management, illegal littering, and on-site mechanical degradation, which can be discriminated based on the properties of the plastics. Film-type plastics with larger sizes are naturally transported from urbanized areas to remote places by wind and river flow despite of relatively low abundance

of the plastics. Foam-type plastics roll with the wind on the ground and flow with water flow along the tributary.

The sampling sites were classified into two clusters by HCA (Fig. 3.7b), which can be distinguished by their plastic composition. Same as PCA, T1–T3 sites near Ulaanbaatar City were assigned to the first cluster. Other sampling sites were assigned to the second cluster. In addition, the Kh (Darkhan City) and O5 (Sukhbaatar City) sites can be discriminated from other sites in the second cluster based on the population density. Statistical multivariate analysis proves that the major plastic waste mainly consisting of MPs concentrated in highly populated areas, while light mega-sized plastics are transported from the urban district to remote places. In addition, micro-PSF and plastic bags or sandbags represented typical plastic waste on river shores of Northern Mongolia with high population densities.

3.3.5 Regional specific plastic "Polystyrene foam (PSF)"

In this study, a relatively high number of foamed plastics, especially micro-sized PSF (Fig. 3.8 and 3.9) was characteristic in the Selenga River system. A variety of PSFs is used for various applications and they generate a very appreciable volume of waste (de Paula et al., 2018). The expanded PSF is frequently employed as packaging for different goods and a heat insulator in buildings and constructions. Unfortunately, as this material is difficult to recycle, their reuse or recycling has little interest for business due to low economic return (Gutiérrez et al., 2013). Owing to frequent consumption and low recycling rate, a relatively high number of expanded PSFs were found in the Selenga River system. High-density distribution was observed on the river shore near Ulaanbaatar City (T1–T3) and that of the Kharaa River near Darkhan City (Kh).

Harsh climate conditions during the long, extremely cold winter season are the main reason for the consumption of foamed plastic material as heat insulator. However, the field survey near Sukhbaatar City (O4 and O5) indicated 2.3 and 0.5 pieces per unit area (100 m^2) of micro- (O4) and macro-sized (O5) PSFs, respectively, probably due to the smaller population density in Sukhbaatar compared with that in Ulaanbaatar and Darkhan. Similarly, only a few pieces of mega- and macro-sized PSFs were observed in the study area at the O3 and Ye sites. Foams were not observed at the study sites O1, O2, and Sh. The lower number of foamed plastics at remote sites suggests that construction in urbanized areas and packaging are the main reasons for PSF occurrence in areas with high population densities, while only several pieces of foamed plastic materials were counted at remote field sites. The

scattering of plastic waste in remote places with low population densities possibly decreases the number of plastic wastes during the transportation from populated zones to river shores.

Several studies highlighted that foamed MPs originate from packaging, fishing gear, and materials for horticulture in river systems and ocean systems (Barnes et al., 2009; Besseling et al., 2017; Faure et al., 2015; Fok et al., 2017; Lee et al., 2013; Moore et al., 2011; Reisser et al., 2013). A number of potentially harmful chemicals identified from the polystyrene plastic material in the environment (Faure et al., 2015; Gambardella et al., 2017; Hüffer et al., 2018; Li et al., 2017). It could be confirmed that these materials were transported chemicals such as POPs (Mato et al., 2001a), including chemicals prohibited to be produced (e.g., poly chlorinated biphenyl) and plastic additives (such as plasticizers, heat stabilizers, pigments, etc.) (Mato et al., 2001a; Talsness et al., 2009), micro-organisms and chemicals (Barnes, 2002) to other locations. The other polymer types of foam materials such as PU foam can also carry heavy metals and other pollutants of the beach sediment of southwestern England (Turner and Lau, 2016). Numerous harmful effects of PSF plastics would be dangerous with respect to food web in aquatic environment. It is necessary to study PSF pieces and understand their behavior in the freshwater environment.

3.4 Conclusion

Various amount of plastic debris found in the river shores of Selenga River system, which indicating that major river system of Mongolia could be one of the pathways of plastic migration in the terrestrial environment. The average number of plastic debris found on the river shores is not comparable to that of previous studies, but the composition of the size fractions and materials are rather similar to that of other studies. Noticeably high amount of PSFs in the study area is characteristic compared to other studies, which reported high contribution of polyethylene films, polypropylene fragments, resin pellets and fibers. Heat insulator and construction materials highly contributed to the PSF debris in the environment. The plastic debris can be differentiated based on the type of materials and the sizes during its transportation.

The distribution of plastic debris is distinctively high around the capital with a high population density. The capital and other towns with high population density are the source of plastics, which are spread to remote places along tributaries by water currents and water level changes. The results demonstrate that low population areas can be polluted by plastic debris through wind and water transports improper waste management. Significant linear

correlations between the size fractions of PSF and polyethylene films indicate that all research sites represent plastics with similar size fractions. The size reduction of these plastics occurs on-site by freeze–thaw processes, solar radiation, and physical fragmentation. The dominance of plastic debris by MPs in aquatic environments reflects the size reduction during their transport.

Predominantly distributed PSFs were considered as a carrier of harmful chemicals in the aquatic ecosystem which arise further concerns relating to their interaction with other synthetic materials including invisible MPs in the environment. The increase in surface area through size reduction enhances the potential to interact other pollutants in the environment. It is necessary to address the aging processes of plastic debris, which increases the surface area and thus raise up the potential as pollutant carrier.

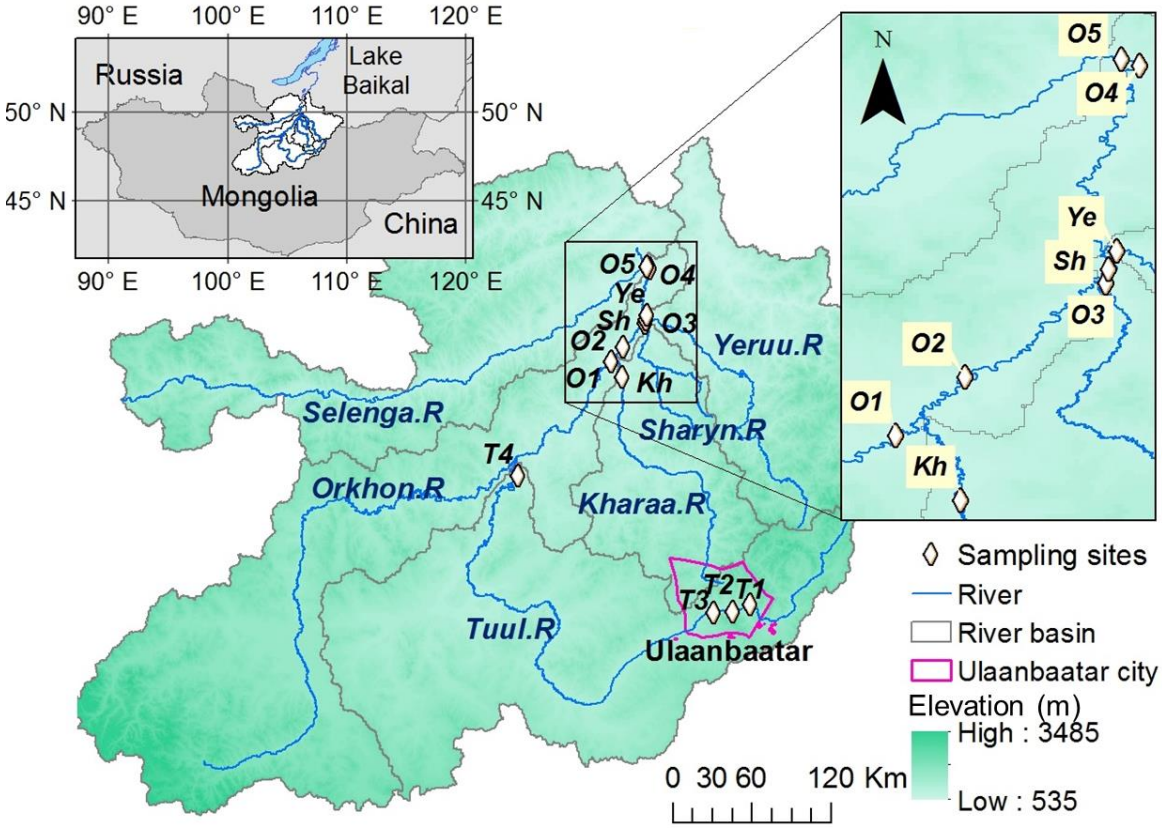

Fig. 3.1 The Selenga River basin in the Mongolian territory and study sites in the basin with sampling points.

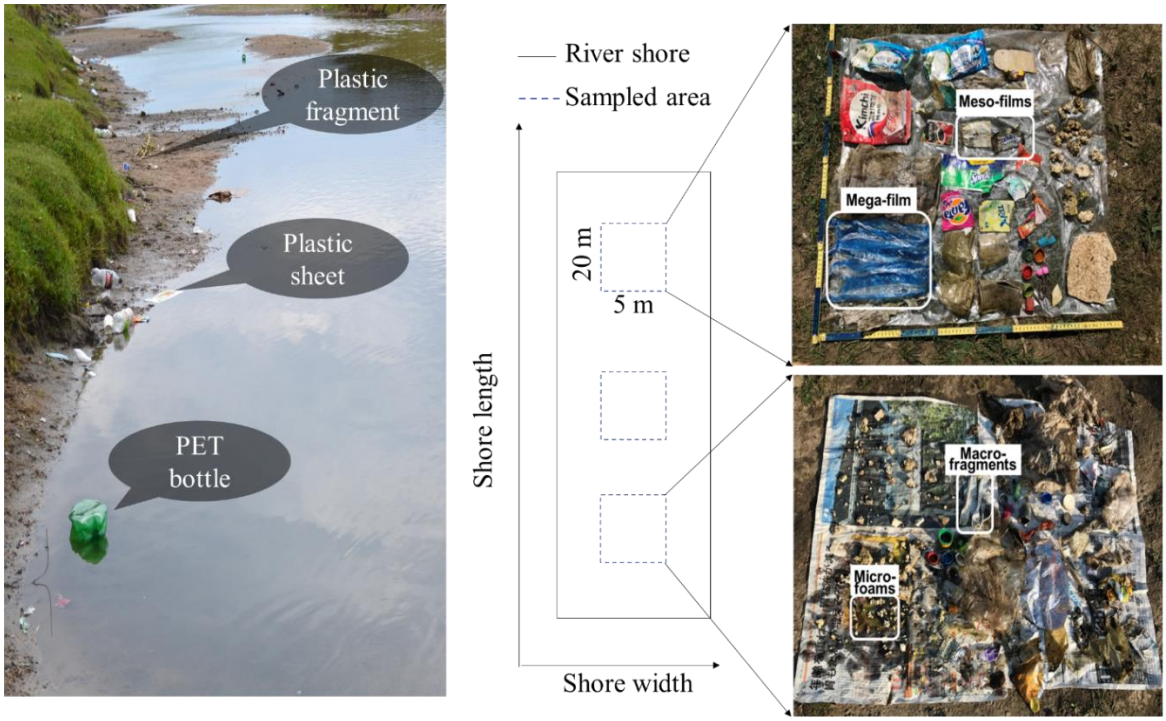

Fig. 3.2 An example of river shore with the sampling technique of shore sampling. Plastic debris collected at the T2 site on the shore of the Tuul River. Mega- and meso-films, macro-fragments and micro-foams marked by white squares in the collected plastic samples.

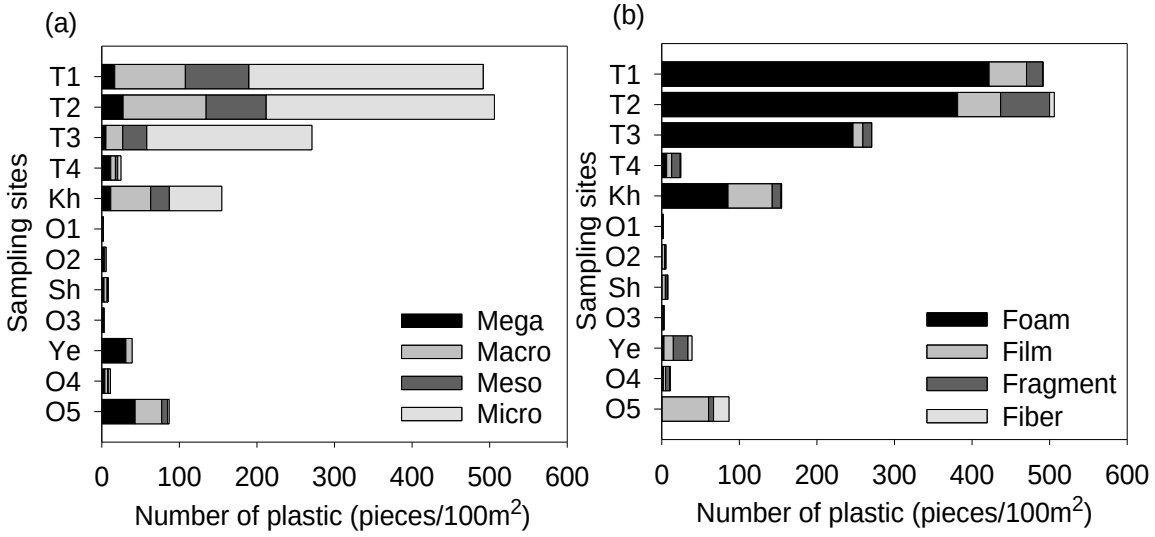

Fig. 3.3 Plastics counted at the study sites. (a) Composition of the size fractions at each site (b) Composition of plastic morphotypes at each site.

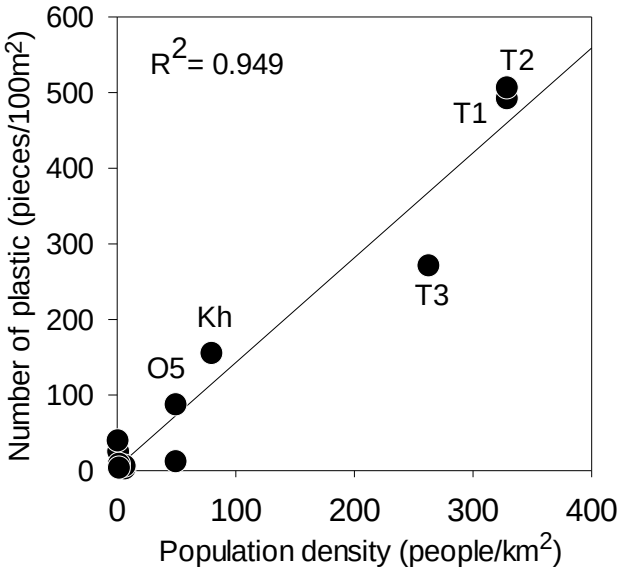

Fig. 3.4 The relationship between the number of plastics in a research area and population density at the town, village or administrative districts including the sampling site.

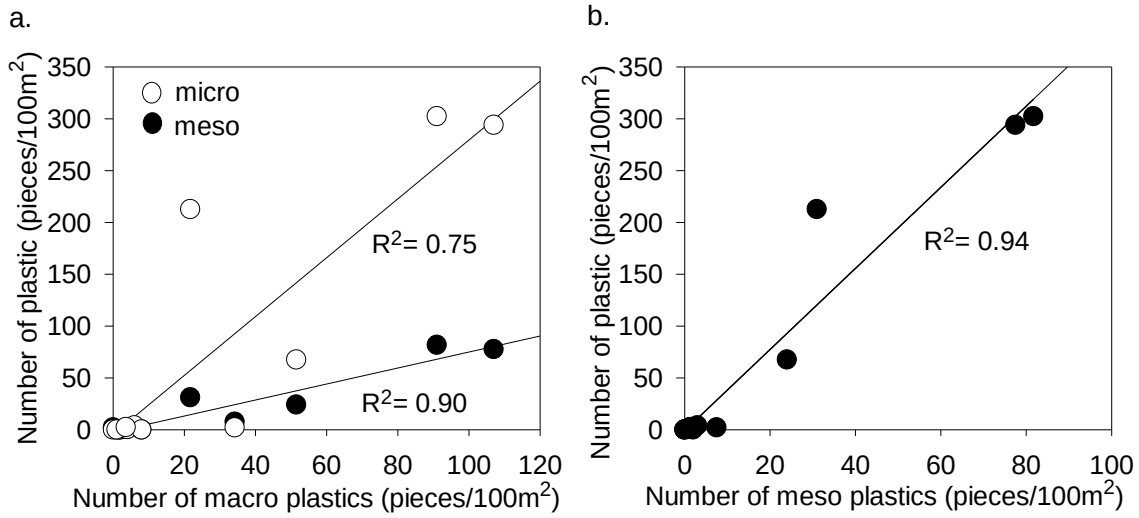

Fig. 3.5 The relationship among the number of plastics in the size fractions. a. Macro- and meso-sized plastics; and macro- and micro-sized plastics; b. Meso- and micro-sized plastics.

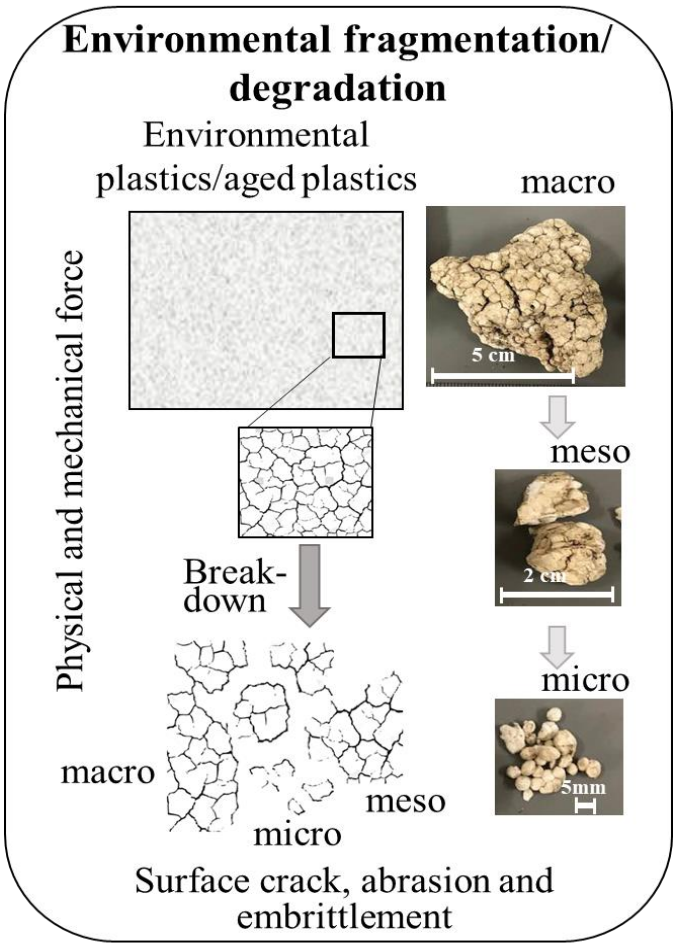

Fig. 3.6 The graphical illustration of fragmentation of plastics with an example of environmental PSFs.

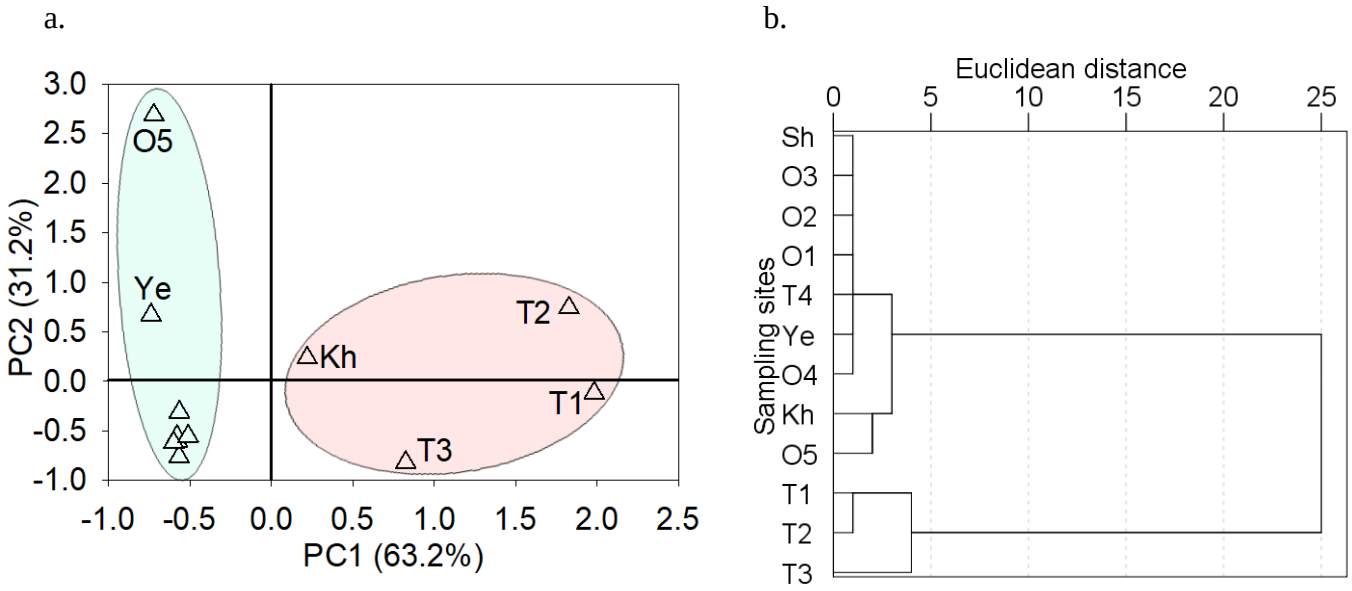

Fig. 3.7 Multivariate statistical analysis. a. Scores plot for the two principal components of sampling sites; b. The HCA of sampling sites based on PCA.

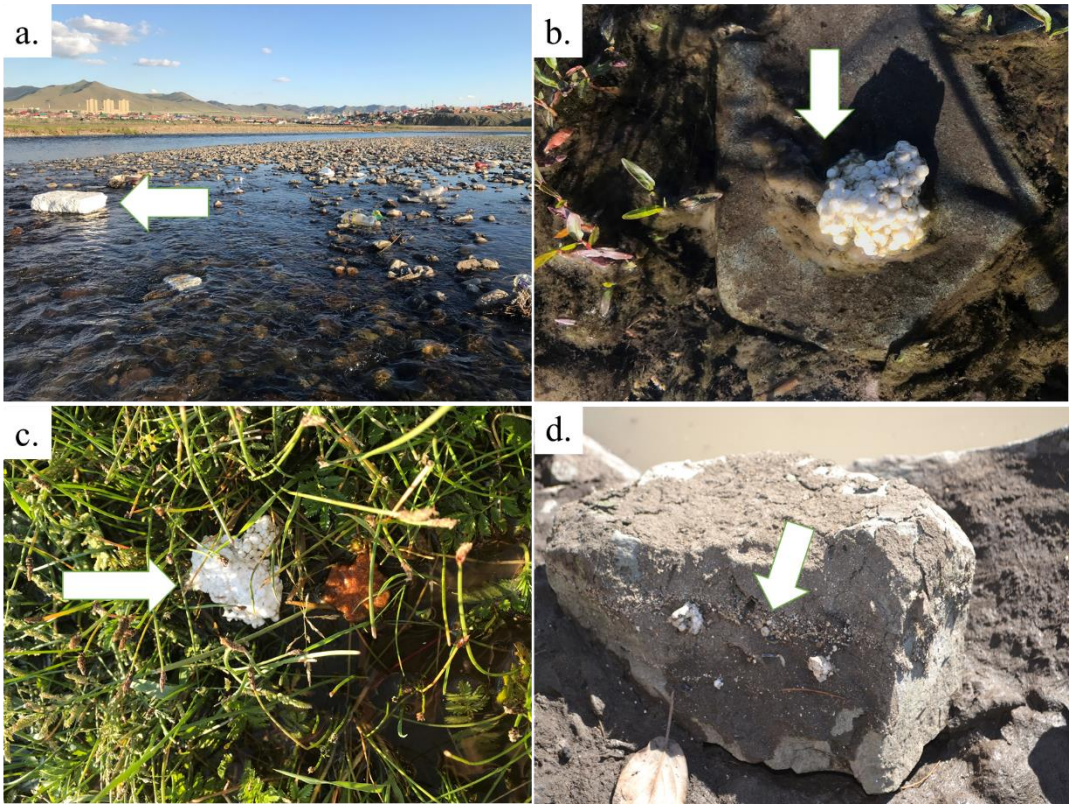

Fig. 3.8 Examples of recorded various sized PSF in the Selenga River system. a. Mega-sized, b. Macro-sized, c. Meso-sized, and d. Micro-sized PSFs on the river shores.

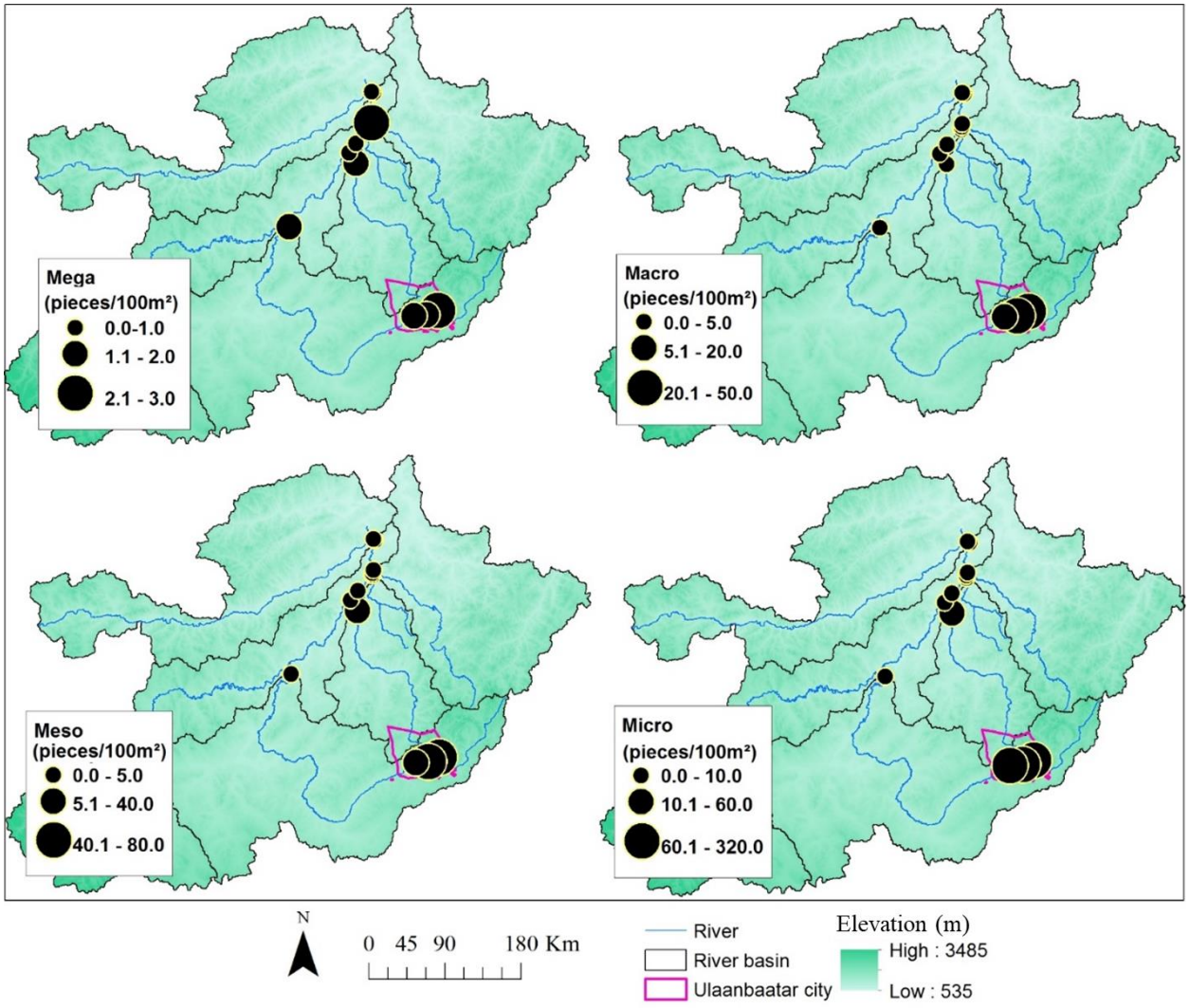

Fig. 3.9 Distribution of PSF according to its size fraction at the sampling sites. The sizes of closed circles are the number of plastic fragments in a surveyed area ($100m^2$).

Table 3.1 Abundance of plastic pieces according to different size fractions, dominant morphotypes and polymer types observed in the research fields.

Size	Average number of pieces/100m^2 ± SD	Dominant type of plastic material (%)	Major polymer type
Mega	3.93 ± 13.73	Film (51.2%)	Polyethylene
Macro	10.88 ± 37.03	Film (43.4%)	Polyethylene
Meso	14.90 ± 30.04	Foam (77.5%)	Polystyrene
Micro	120.14 ± 121.49	Foam (99.0%)	Polystyrene

SD: standard deviation

Table 3.2 Correlation matrix of film and foamed plastic material.

	Mega	**Macro**	**Meso**	**Micro**	**Pop.den.**
a. Film					
Mega	1				
Macro	0.81**	1			
Meso	0.82**	0.89***	1		
Micro	0.57	0.41	0.77**	1	
[1]Pop.den.	0.45	0.51	0.71*	0.60*	1
b. Foam					
Mega	1				
Macro	0.30	1			
Meso	0.29	0.98***	1		
Micro	0.27	0.92**	0.97***	1	
Pop.den.	0.22	0.90**	0.95**	0.99***	1

Pop.den. population density

* p<0.05; ** p<0.01; *** p<0.001

Table 3.3 Extracted components of PCA.

	Component	
	1	**2**
Mega	0.152	0.943*
Macro	0.897*	0.375
Meso	0.984*	0.136
Micro	0.990*	0.021
Film	0.574	0.725
Fiber	–0.114	0.958*
Foam	0.993*	0.030
Pop.den.	0.973*	0.072

Pop.den. population density

*Significant values

CHAPTER 4

Aging of polystyrene foams and properties of adhered microplastics in the urban river

4.1 Introduction

Polystyrene is one of the most commonly produced and consumed polymer materials in the world. An extensive use of polystyrene materials promotes scattering of these plastic wastes as marine (Didier et al., 2017; Fok et al., 2017) and freshwater (Moore et al., 2011) debris due to low level of waste management with respect to dominance of single-use products. Owing to ubiquitous distribution and interaction with other pollutants in the environment, aging of polystyrene plastics and their behavior could be important information to understand plastic pollution in the aquatic environment.

Plastics have various durability with different degradation rates based on the source of plastics, types of polymerization, chemical additives, and environmental conditions (Gewert et al., 2015). For detailed information on the surface property of plastics, several signs such as yellowing, cracking, embrittlement etc., can be observed in the plastics as indicator of degradation. Instead of physical changes of plastics, the chemical changes accompanying the formation of oxygen-containing functional groups and the decrease in molecular mass are other important signs of plastic degradation (Bandow et al., 2017). Although the durability of plastic has been estimated in the range of tens to hundreds of years (Barnes et al., 2009), aging of environmental plastics can be characterized by different degrees of plastic resistance to various physical (environmental) conditions. For example, acrylonitrile butadiene styrene and polystyrene plastics have poor resistance against UV light, and these plastics can be significantly damaged by environmental factors compared with other polymer materials (Castillo et al., 2016; Weinstein et al., 2016). The overall light-initiated process in the presence of oxygen is generally referred to as oxidative photodegradation or photooxidation. Using the photooxidation status, it is possible to evaluate the relative aging by carbonyl index of polymer materials because UV radiation promotes production of carbonyl group on the polymer structure. On the other hand, degradation of plastics increases their sorption potential of pollutant chemicals through microcracks and oxygen containing

functional groups, resulting in the ingestion of toxic chemicals by aquatic organisms (Gao et al., 2019; Vroom et al., 2017).

Following the travel and aging of plastics in the environment, plastics not only degrade and fragment but also aggregate in the environment. The plastics photodegradation and the formation of aggregates have important effects on the environmental fate and toxicity of plastics. Despite various microplastic (MP) studies, a comprehensive understanding of environmental degradation and interactions between aged plastics and other MPs is still rather limited (Hüffer et al., 2018; Lee et al., 2014). Since plastics interact with other pollutants in the environment, polystyrene foam (PSF) which is predominantly found on the river shores of Selenga River system in Mongolia, considered as a carrier of invisible MPs through formed aggregates during their travel in the environment. To understand the characterization of adhered MPs, this chapter focused on the distribution of PSFs and their interactions with adhered MPs. The photooxidation status is also evaluated for PSF plastics. The properties of aggregates can be useful for understanding the behavior of degraded plastics in freshwater ecosystems. The aims of this chapter are as follows: 1) to understand the behavior of plastics, particularly PSF, in the urban river in Ulaanbaatar City, 2) to identify the photooxidation status of PSF using carbonyl index, and 3) to determine the amount of MPs that formed aggregates with PSFs and characterize them. The importance of this chapter is a consideration of plastic-plastic aggregates based on aging behavior in the aquatic environment in order to characterize the fate of environmental plastic debris.

4.2 Materials and methods

4.2.1 Tuul River and waste management in Ulaanbaatar

The river chosen for the quantification and evaluation of PSF was Tuul River (Fig. 4.1) running in the center of Ulaanbaatar because large amount of PSFs with various sizes were detected from point and non-point sources in the urbanized area as mentioned in the Chapter 3. The Tuul River has a watershed area of 49,840 km^2 and it is one of the important headwaters of Selenga River system (Baljinnyam et al., 2014). The Tuul River originates from the Khangai mountains and passes through Ulaanbaatar City before meet the Orkhon River. The river flows through the steppe region (80% of the river basin area) which mostly used for pastureland. Forests (7%), hay and croplands (3%), and urban areas (1%) are the other land cover categories of the river basin (Pietroń et al., 2017). The annual precipitation

ranges from 253 to 275 mm, and the long-term annual average flow of the Tuul River is about 25.6 $m^3 s^{-1}$ at Ulaanbaatar hydrological station (Sukhbaatar et al., 2016).

The upstream of the river supplies the primary drinking water for Ulaanbaatar citizens, while the industries use large amount of water from the downstream of the Tuul River and adjacent aquifer systems (Pietroń et al., 2017). Additionally, direct input of sewage water and effluent from the WWTP have been major reasons for water pollution of Tuul River (Altansukh et al., 2012). The central sewage canals and excessive wastewater released from the WWTP directly discharged into the main channel of Tuul River in the Ulaanbaatar region (Fig. 4.1) for this half century. The wastewater treatment capacity of the WWTPs have been exceeded by actually released wastewater during last 50 years due to population increase and lack of proper renovation and maintenance of the plant. It has been reported that the efficiency of the wastewater treatment dropped to 71% in 2002 and further to 66% in 2003, with respect to the total released wastewater. Recently, the total amount of daily wastewater was over 160 to 170 thousand cubic meters, which is three times higher than the capacity of the WWTP (Altansukh and Davaa, 2011).

Besides the effluent discharge into Tuul River, municipal solid waste issues have been another concern of pollution along the Tuul River. Although Environmental Protection law of Mongolia (1995) placed responsibilities on the various administrative divisions and sets out the rights of citizens concerning waste management services, the released waste from the urban area only managed by collection, transportation, storage and landfill methodology without proper classification and regulated re-cycling processes (Byamba and Ishikawa, 2017). The improper waste managements at the final waste treatment station in Ulaanbaatar and industrial and mining activities can be the sources of pollutants, which are exacerbated by further waste discharges to downstream water bodies.

After the democratic movement in the 1990s, a rapid population migration from rural to Ulaanbaatar areas promoted the development of urban infrastructures in this region. However, the capacity of the infrastructures has been insufficient to accommodate the excessive wastewater requirement due to the rapid increase in the daily consumption of water and consumer products. The release of municipal solid wastes has annually increased since the 1990s from approximately 550 thousand tons to 969.1 thousand tons in 2015 (Asian Development Bank, 2008; Byamba and Ishikawa, 2017), resulting in the creation and expansion of dumping sites in Ulaanbaatar to fulfil the overwhelming requirement from the increasing population. Unfortunately, those dumping sites were constructed without any

covers and fences, leading to the inadvertent release of plastic wastes by wind and rainwater along the small tributaries to the Tuul River.

4.2.2 Site description and sampling

In this study, six sampling sites were selected along the shores of the Tuul River in March 2018 to investigate distribution of plastics particularly PSF debris depending on their sizes in the field (Fig. 4.1). As highlighted in the Chapter 3, environmental PSFs were found as dominant plastics of the micro-size fraction. Sampling sites were selected based on geographical background from upstream to downstream of river. The sampling site, S1, was located at the upper stream of the Tuul River in the eastern part of the area of Ulaanbaatar city with a low-density population. The S2, S3 and S4 sites were located at the middle part of the city with concentrated urban districts. These central three sites were affected by sewage water through central drainage canals from the highly populated area of Ulaanbaatar. The S5 and S6 sites were located at the downstream of the river with relatively low population density. In addition, sampling sites S3 and S5 have been influenced by the city sewage from the WWTPs. The river has several small tributaries from the northern part of Ulaanbaatar city. Small tributaries have washed out unexpected wastes, including plastics from the city district, to the mainstream during the rainy and snow-melting seasons.

The sampling was the same way as described in the Chapter 3. Briefly, plastic samples collected from the river shores with selected 100 m^2 area. The triplicate quadrates were set on the river shores in each sampling site and all visible plastics were collected by hand-sorting technique. Smaller sized and large number of plastics on the shore were carefully collected from the accumulated spot. The collected plastic pieces were identified by their sizes (macro: 100 – 20 mm; meso: 5 – 20 mm; and micro: <5 mm) (Barnes et al., 2009; Suaria and Aliani, 2014). The lower limit of MPs was 1.25 (±0.4) mm due to their visible size and collectable by hand. Although mega-sized PSF (>100 mm) were found at the fields, further analyses to evaluate the surface carbonyl index and the aggregation status with MPs were not conducted for them because of their uncommon distribution at the research sites.

4.2.3 Sample treatment and analysis for degradation status of PSF

Polystyrene foam samples were evaluated in terms of their chemical structural properties. From each quadrate, a counted number of macro-, meso-, and micro-sized PSF

subsamples with triplicates (n = 162) digested to remove natural organic matter covered onto PSF surface. At first, the subsample pieces of PSF were counted. The digestion of organic matter was carried out by the Fenton reagent, in a conical beaker on the hot plate (75°C). Figure 4.2 have shown the PSF samples before and after digestion of organic matter using a digestion mixture of Fenton reagent. From the photographs in Fig. 4.2, it is clearly shown that biofilm was removed from the surface of PSF samples. The organic matter digestion was conducted 24 – 48h, including repetition of the Fenton regent addition and settling time at the room temperature. The conical beaker was covered with a watch glass and the complete removal of organic substances confirmed by naked eyes. The conical beaker was placed in an ultrasonic bath for several minutes to separate the PSF complex. Contents in the conical beaker were passed through a Whatman glass microfiber filter (GF-F, Whatman, Co. Ltd) to collect the PSF particles and separate the adhered particles under vacuum condition. PSF samples were collected on a glass petri dish using a tweezer and then dried at room temperature in a desiccator for several days to avoid airborne contamination. The digestion was also applied to reference PSF particles to confirm any effects from the Fenton reagent on the PSF surface. The glass microfiber filter with retentates was kept for further analysis to identify the retentate particles for MP determination. The surface areas of subsamples in macro-, meso-, and micro-sized PSFs were calculated by direct measurement of three dimensions (length, width, and height). In the case of micro-sized PSF, the diameter of a single PSF particle was determined using a micrometer caliper. The spherical surface area was calculated from the diameter of the particle.

To identify the chemical structure and surface oxidation status of PSFs, the recovered samples were analyzed using a micro- FTIR (Shimadzu, Kyoto, Japan). A target area in a microscopic view was fixed at several micrometers with 100 µm width on a surface of the PSF followed by scanning in a range of 4000 cm^{-1} to 500 cm^{-1} by micro-FTIR as its reflection mode. Spectra were obtained after 100 scans. From the surface of a PSF, three different targeted areas were scanned to confirm the spectra of the PSF sample. IR spectra were also obtained from the digested reference PSF in the same manner.

In this study, we adopted carbonyl index to identify the oxidation status of the surface of PSF, which can be reflected the absorbance of carbonyl moieties on the surface of PSF (Botelho et al., 2004; Mylläri et al., 2015; Shi et al., 2019). The carbonyl was calculated using the equation (*4.1*) as follows:

$$Carbonyl\ index = \frac{A_{-CO-}}{A_{-CH_2-}} \qquad (4.1)$$

where A_{-CO-} is the absorption from carbonyl group observed at 1732 cm^{-1} and A_{-CH_2-} is the absorption from methylene group observed at 2850 cm^{-1}. The relative level of surface oxidation was also expressed as "low", "moderate" and "high" whereby their carbonyl index corresponding values were defined between 0.00 and 0.20, 0.21 and 0.40, and higher than 0.41.

4.2.4 Quantification and identification of adhered microplastics (MPs)

The number of MPs adhered onto PSF samples collected on a glass microfiber filter was calculated using the theoretical area of the filter and the average number of MPs in a fixed area of square (12 mm^2 of area), which was randomly selected under a digital microscope with 100 times magnification (VH-7000, Keyence, Japan). The number of MP items on a piece of PSF (macro, meso, and micro) was estimated from the total number of MP items on the filter after digestion of PSFs for further discussion. Experimental blanks from the reference PSFs were performed using the digital microscope to evaluate any possible airborne contaminations during the experiment; as a result, additional MP contamination was not detected during the experiment. Simultaneously, individual items of the MPs adhered onto PSF were categorized into four morphotypes (fiber, film, foam, and fragment) from the shapes (Davis and Murphy, 2015) and morphologies in the microscopic views using the digital microscope. Under a microscopic view of the micro-FTIR, a square frame was fixed targeting the MP which was separated from PSF to obtain IR spectra with an identifiable signal/noise ratio to characterize their chemical origins. The smallest square enough to obtain spectra was 10 × 5 μm. The operation condition for the FTIR analysis was the same as that conducted on the surface of PSF. The spectra of the suspected MP particles were compared to the Aldrich library of FTIR spectra and the polymer types were determined (Pouchert, 1985). Statistical analysis was performed using SigmaPlot V12.0 (Systat Software Inc. USA) and SPSS Statistics V22.0 (IBM Inc, USA).

4.3 Results and discussion

4.3.1 Abundance and distribution of PSFs along the Tuul River

The composition of size fractions was different in distinct morphotypes in these research areas (Table 4.1). Foamed plastics were dominant in most of the size fractions, except for the mega plastics (Table 4.1). The highest distribution density of PSF was observed at S4 where the confluence of sewer discharges from the city center (Fig. 4.3). Micro-sized PSF was the dominant piece of plastics at all sites in Tuul River (Fig. 4.3). The dominant type of MPs was characteristic in different research regions. For example, the Atoyac River basin in Mexico was dominated by films and fragments of MPs (Shruti et al., 2019), whereas fiber-type MPs were the most abundant in the South Yellow Sea in China (Wang et al., 2019b). Micro-sized PSF was also the common dominating MPs in several regions (Moore et al., 2011; Rodrigues et al., 2018a). In addition, Kang et al., (2015) reported that a relatively high abundance of styrofoam has also been originated from aquaculture materials in the Nakdong River mouth in Korea. Given their small size and large amount in the aquatic environments, MPs have led to a further fatal impact on the aquatic ecosystems (Besseling et al., 2015; Duncan et al., 2019; Silva-Cavalcanti et al., 2017).

The linear relationships between mega- and macro-sized ($p < 0.01$ and $r^2 = 0.80$), macro- and meso-sized ($p < 0.01$ and $r^2 = 0.94$), and meso- and micro-sized ($p < 0.01$ and $r^2 = 0.52$) PSFs were a proof of on-site fragmentation at the field (Fig. 4.4). In this study, micro-sized PSFs were usually collected as single spheres, indicating that the fragmentation of technically aggregated PSFs had already progressed after their release into the environment. On the other hand, PSFs were linearly distributed along the river, probably due to the effect of seasonal or annual water level changes (Fig. 4.5). The distribution and abundance of plastics could be attributed to the types of plastics used as well as the high population density. However, the growth of microorganisms on the micro PSF particle altered its specific gravity to heavier, resulting that the PSF covered with biofilm can sink to the bottom of river and further accumulate on the shore (Carson et al., 2013; Cole et al., 2011). The PSFs were commonly found as foams with brownish color (Fig. 4.2), mainly covered with organic substances, oxides, and/or fine minerals during their transportation. The aged plastics in the environment changed their physical properties, such as specific gravity, surface roughness, color and surface structure; hence, the aged PSFs could be dispersed in the river water

followed by sedimentation as river bottom sediments and/or river shore sediments by seasonal changes in water levels.

Given attention to the polystyrene plastic degradation and fragmentation in the environment, the accumulation of styrene oligomers that originated from polystyrene plastics in the aquatic environment have been reported by Amamiya et al., 2019; Kwon et al., (2018, 2015, 2014) and Saido et al., 2014. The reported studies on the styrene oligomers in the environment highlight that styrene oligomers are leached from the discarded polystyrene particles when incomplete polymerization occurred during polystyrene manufacturing. Figure 4.6 shows the diagram of leaching process of styrene oligomers from the discarded polystyrene in the beach environment (Kwon et al., 2015) which indicates that environmentally distributed plastic materials, particularly polystyrene plastics, have high potential to degrade, fragment and release the chemicals into surrounding environment.

4.3.2 Surface oxidation of PSF

The surface oxidation of each of macro-, meso-, and micro-sized PSFs was evaluated using carbonyl index to monitor the photooxidation status of PSF debris. The carbonyl index of PSFs ranging between 0.00 and 1.09 indicates diverse stages of photooxidation status on the surface of all the size fractions of PSFs after their alteration of the surface (Fig. 4.7). Additionally, the reference PSF was evaluated the surface oxidation status using the carbonyl index in order to find any influence from the digestion mixture. Figure 4.8 shown that there is no significant effect from the Fenton reagent and sonication on the PSF samples (carbonyl index = 0.00). Therefore, PSFs with high values of carbonyl index were the results of photooxidation after exposure of new face of PSF to solar radiation in the environment. The range of carbonyl index suggests that differences in progress of photooxidation during the exposure of the new surface after fragmentation. Accompanying with the residential time at the sites where PSF debris reached, the carbonyl index values of PSFs can be vary due to surface coverage by organic matter (biofilms) in the environment. The wide range of carbonyl index can be a good indicator to evaluate the behavior of PSFs on the river shore rather than the number of distributed PSFs on the site (Fig. 4.7).

The highest carbonyl indices in different size fractions were 1.09 (macro), 1.02 (meso), and 1.00 (micro) observed at S6, S5, and S3 sampling sites, respectively. In addition, downstream sampling locations were dominated by highly oxidized PSFs with high carbonyl index (Fig. 4.7). The wide range of carbonyl index in micro-sized PSFs was characteristic at

S3 sampling site, where it has been influenced by discharges from the city sewage canals. The wide distribution of carbonyl index in micro-sized PSFs expressed by a broad peak in Fig. 4.7c was characteristic as compared to that of macro- and meso-sized PSFs. On-site fragmentation processes allowed PSFs to increase their specific surface areas exposing new surfaces to the air, resulting in the concurrent oxidization of surfaces and exposure of the new surfaces. As a result, secondary released micro-sized PSFs have complex stages of oxidation consisting of highly oxidized old surfaces and new surfaces after their fragmentation. On the contrary, aged micro-sized PSFs which has kept their surface exposed to air for a longer time, results in high carbonyl index due to continuous oxidation in the environment.

In addition, several studies in a laboratory scale tried to evaluate changes in surface properties of polystyrene plastics through photodegradation leading to adsorption of toxic chemicals or polymers (Botelho et al., 2004; Liu et al., 2019a; Shi et al., 2019). These studies observed carbonyl index values on polystyrene polymers through artificial aging which are two to three times higher than those of our results. Newly generated surfaces also affected the averaged photooxidation status of the PSF particles due to the complexity of the surface including both newly exposed surfaces, aged surfaces and biofouling surfaces.

The degree of oxidation was classified into three oxidation stages in this study: i. low (carbonyl index: 0.00 – 0.20), ii. moderate (carbonyl index: 0.21 – 0.40), and iii. high (carbonyl index: >0.40) (Fig. 4.9). The low degree of carbonyl index was mainly accounted for 65% in macro-, 63% in meso- and 57% in micro-sized fractions, respectively. The moderate stage was common in the meso-sized fraction (34%), whereas the high stage of carbonyl index was observed in micro-sized fractions (15%) by photooxidation leading to chain scission of chemical bonds in the polystyrene polymers. Initial degradation by UV radiation can promote the degradation stages of PSFs with lower carbonyl index irrespective of environmental conditions. However, changes in chemical and physical properties of polymers could also be other indicators to evaluate the degradation stages of plastics because additives such as UV stabilizer, flame retardants, antioxidants and, plasticizers could influence photooxidation of polymers (Hermabessiere et al., 2017). Polystyrene foam consisting of porous structure could be broken down easier by mechanical and physical forces on PSFs. Furthermore, highly oxidized or aged MPs could be easily broken down into invisible micro-sized fractions, increasing their adsorption potential, ingestion, and bioaccumulation owing to their larger specific surface area (Gambardella et al., 2017).

Plastic debris undergo several complex processes during their trip, such as desorption of chemical additives from polystyrene particles (Coffin et al., 2019; Jang et al., 2017; Jeannerat et al., 2016) and adsorption of organic pollutants on the surface of polystyrene particles (Hüffer et al., 2018; Li et al., 2017). The possibility of desorption and adsorption of contaminants on plastic debris has been reported in several studies of aged polystyrene MPs. Aged polystyrene MPs have lower potential to adsorb chemicals compared with newly released polystyrene MPs (Hüffer et al., 2018). On the other hand, beached PSF has higher oxytetracycline adsorption compared with any pristine PSF (Zhang et al., 2018a). This result provides a further assumption that a higher number of MPs with the lower stage of surface oxidation might have an adverse effect on aquatic ecosystems by interaction and accumulation of contaminants.

4.3.3 Properties of adhered MPs

Microplastics extracted from the surface of macro-, meso-, and micro-sized PSFs have been observed under the microscope (Fig. 4.10). The abundance of adhered MPs ranged from 5 to 141 items on a piece of PSF. It is important to note that adhered MPs were ubiquitously detected from the surface of PSFs in this urban river environment. A sorption mechanism of plastics with organic pollutants (Liu et al., 2019b; Wang et al., 2018), concentrations of adsorbate (organic pollutants and trace metals) (Bakir et al., 2012; Prunier et al., 2019), and accumulation of microorganisms on the plastics (Jin et al., 2018; McCormick et al., 2014) have been reported in the marine and freshwater ecosystems, whereas there is no particular study relating to the interactions between plastics. Since the concentration ranges of MPs were detected with different orders of magnitude in the aquatic systems all over the world (Anderson et al., 2016b; Rezania et al., 2018), the effects of MPs on the distribution and behavior of adhered MPs in the aquatic environment would be necessary to predict the following fate of MPs.

The average numbers of adhered MPs were 102, 97, and 14 items on a piece of macro-, meso-, and micro-sized PSFs, respectively. Macro- and meso-sized PSFs with many cracks, holes, and disturbed surfaces have higher potential to interact to invisible MPs with sizes below 1 mm compared with spherical micro-sized PSF particles. On the other hand, the interaction between MPs and organic compounds was driven by physical–chemical processes based on the surface area, pore volume, and hydrophobic bonding, which overall would influence their potential of interaction (Hüffer and Hofmann, 2016; Wang et al., 2018).

Using the calculated surface areas, adhered MPs in a unit surface area were determined in macro-, meso-, and micro-sized PSFs (Fig. 4.11). The adhered MPs in a unit of surface area were 3, 15, and 20 items in macro-, meso-, and micro-sized PSFs, respectively. The higher distribution range of adhered MPs on the surface of micro- and meso-sized PSFs was characterized by their longer residential time to interact with other particles and contaminants in the environment.

There have been published researches discussed about interactions between plastic debris and chemical compounds. Invisible micro- and nano-sized polystyrene particles interacted with hydrophobic compounds through the π–π electron-donor–acceptor interaction, which was highly dependent on the specific surface area and particle sizes (Wang et al., 2019a). The majority of aged MPs had rough surfaces in the aquatic environment (Hüffer et al., 2018; Liu et al., 2019a). Those defects, such as microcracks, fractures, pits, and adhered oxides, increased the potential of interaction between aged MPs and hydrophobic compounds (Cooper and Corcoran, 2010; Jiang et al., 2019). Changes in the surface physical-chemical properties and the hydrophobicity of the plastics were two possible drivers for mechanism of aggregation probably observed in this study, although the aggregation status of MPs was altered by presence of humic substance (Liu et al., 2019a; Xu et al., 2018). These phenomena suggest that the interaction between visible and invisible plastics could be attributed to various factors occurred on surface of PSF plastic debris (aging, fragmentation leading to new faces, and biofilm cover). On the other hand, micro and nano-sized polystyrene beads have been used to understand dynamics of plastic aggregation in laboratory studies (Cai et al., 2018; Li et al., 2018b; Saavedra et al., 2019). Reported studies highlighted that the aggregates consisting of prepared micro and nano-sized polystyrene beads were formed in the controlled laboratory condition. Additionally, Li et al., (2018b) mentioned that polystyrene MPs are likely to aggregate in the marine environment because of influences of monovalent (i.e., NaCl, $NaNO_3$ and KNO_3) and divalent (i.e., $CaCl_2$ and $BaCl_2$) electrolytes with and without humic acids. Furthermore, Cai et al., (2018) mentioned that natural organic matter had negligible effects on aggregation of polystyrene and nanoplastics in prepared NaCl and $CaCl_2$ solutions.

Morphotypes of adhered MPs found on the surface of the three-sized PSFs did not show any specific distribution at all locations (Table 4.2). Adhered foams, films, fragments and fibers occupied 22%, 21%, 28% and 29% of the total abundance of adhered MPs, respectively. The evenly distributed morphotypes of adhered MPs indicated that PSFs had no unique selectivity to MP morphotypes in the environment. However, there were various

chemical structures of fibers (polyacryl, polyester, and polyamide), films (polyethylene), fragments (polypropylene, polycarbonate, polyvinyl chloride) and foams (PS) that were identified using the micro-FTIR (Fig. 4.12). These types of polymer materials were relatively common plastics used in many industrial sectors and in our daily life (PlasticsEurope, 2018). The polymer composition of adhered MPs onto PSFs were determined (Fig. 4.13A). Polyamide (20.2%), polyester (10.9%) and polyacryl (3.6%) polymers mainly detected as a fiber type of plastics are dominantly adhered onto all three sized PSFs (Fig. 4.13B). The dominance of fibers is attributed to confluence of WWTP effluent to the Tuul River. Foamed type of adhered MPs (polystyrene) were largely found in the surface of macro-sized PSFs compared to meso- and micro- sized PSFs with the rate of 26.2%, 17.1% and 11.0%, respectively. This rate might be related to the surface areas of PSFs, suggesting that fragmented invisible PSFs subsequently interact with parent PSFs through hydrophobic bonding. This is probably one of the major mechanisms to form plastic aggregates after their fragmentation. Furthermore, the percentage of unknown polymer types have increased in the detected invisible MPs from macro-sized PSFs to micro-sized PSFs. High percentage of unknown plastics (20.5%) in the micro-sized PSFs demonstrate that consequences of technical limitation (Li et al., 2018a) to analyze the smaller sized invisible plastic particles with low resolution spectra. Fragmented hard plastics such as PVC and polycarbonate were also existed on the surface of all three PSF fractions (Fig. 4.13B). Difference in the composition of invisible plastic particles could depend on preferential interaction between surfaces of PSFs and invisible polymers as well as abundance of each polymer in the environment. The high rate of fibers and polystyrene in detected invisible MPs was mainly the latter reason.

The rapid expansion of the urban area with the insufficient sewer system in the subcentral residential districts and the growth of numerous textile industries in the Ulaanbaatar area were considered as the major sources of invisible fiber and fragments in the Tuul River system. In addition, the abundance of secondary micro-sized fibers and fragments were reported in the river system due to wastewater supply from the textile industries and the WWTPs (Lares et al., 2018; Salvador et al., 2017). The invisible MPs were not only transported by direct water flow but also carried by flowing MPs through their aggregation formation.

Based on the obtained results in this research, PSFs were recognized as active carrier of various types of invisible MPs in the aquatic environment. The most significant understanding from the aggregates of visible and invisible plastics was that the concentration

of adhered particles was highly influenced by larger specific surface areas and surface properties of PSF materials. However, the PSF plastic material had a unique property to interact various types of invisible MPs from the environment. This study shows the probability of aggregation of invisible plastics in the urban river environment, which also highlights the importance of research in monitoring the adhered MPs in various types of aquatic environments because PSF debris and other MPs have been reported as common plastic debris in marine and freshwater environments. The present findings raise concern about the widespread nature of marine plastic pollution, indicating that plastic-free ocean environments are increasing rare.

4.4 Conclusion

In this study, the distribution and characterization of PSFs on the river shore were evaluated, and the MPs adhered onto the surface of PSFs were also examined. The PSF plastics collected predominantly at the Tuul River basin featured specific characterizations and broad distribution pathway from the released source to the river environment. The behavior of PSFs and the distribution along the river shore should be affected by both environmental and anthropogenic factors. Seasonal changes in water level could leave floating plastic debris on the shore, which could again float on the water surface followed by their downward transportation. The potential of transporting the scattered plastic debris on the river shore could be estimated by determining their distribution density in a unit area of the shore.

In addition, this study provides new insights into MPs in aquatic ecosystem which is variety of secondary sourced MPs actively interact with PSFs and travel together in the outdoor environment. The PSF plastic debris also have a high potential to carry invisible MPs through their aggregation process. The aggregation status of plastic debris could be evaluated by their number. Those measures in monitoring their activity would be valuable to evaluate the pollution status of the river. Afloat, sedimentation and re-suspension with seasonal changes in the hydrological condition of the river can characterize the river to model the behavior of the plastic debris and adhered MPs. Further monitoring of municipal activity and/or population density as robust controlling factors is useful to evaluate the distribution density of plastic debris. The cooperation of government or environmental non-governmental organizations is essential in the future to model the plastic debris behavior.

Moreover, aging is considered the significant feature of interaction between plastics, while mechanism of aggregation is difficult to figure out due to interface of natural organic substances. The aging status of plastic debris is also necessary because highly degraded plastic debris with cracks and coverage of organic substances had a higher potential to carry adhered MPs. The physical status of the plastic debris surface was also an important factor for the aggregates of MPs and PSFs. However, the interaction between plastic debris and adhered MPs was not selective to any specific MPs. All types of plastics were almost evenly adhered onto plastic debris, probably mainly due to the hydrophobic bonding between plastics in the aquatic environment. Specific distribution and abundance of plastic debris occupied by PSF were attributed to the characteristics of plastic consumption in the study region. A similar composition of invisible adhered MPs indicated an autonomous interaction with other plastic particles followed by the formation of plastic aggregates. These plastic aggregates seem to occur in all of the river environments during the movement of plastic debris. From the understanding of this study, MPs undergo aggregation upon release to the aquatic environments, which is one of the critical factors that ultimately controls their environmental fate and ecological risks. Understanding the formation and behavior of the aggregates could be important to evaluate the fate of plastic debris and assess the potential risks to the aquatic ecosystems; thus, research on plastic debris should be conducted considering the aggregation. It is urgently necessary to focus on the spatial and temporal variations in the surface characters of plastic debris, plastic aging, and their aggregates to develop a further understanding on food chains through the plastic intake by organisms in the marine and freshwater ecosystems.

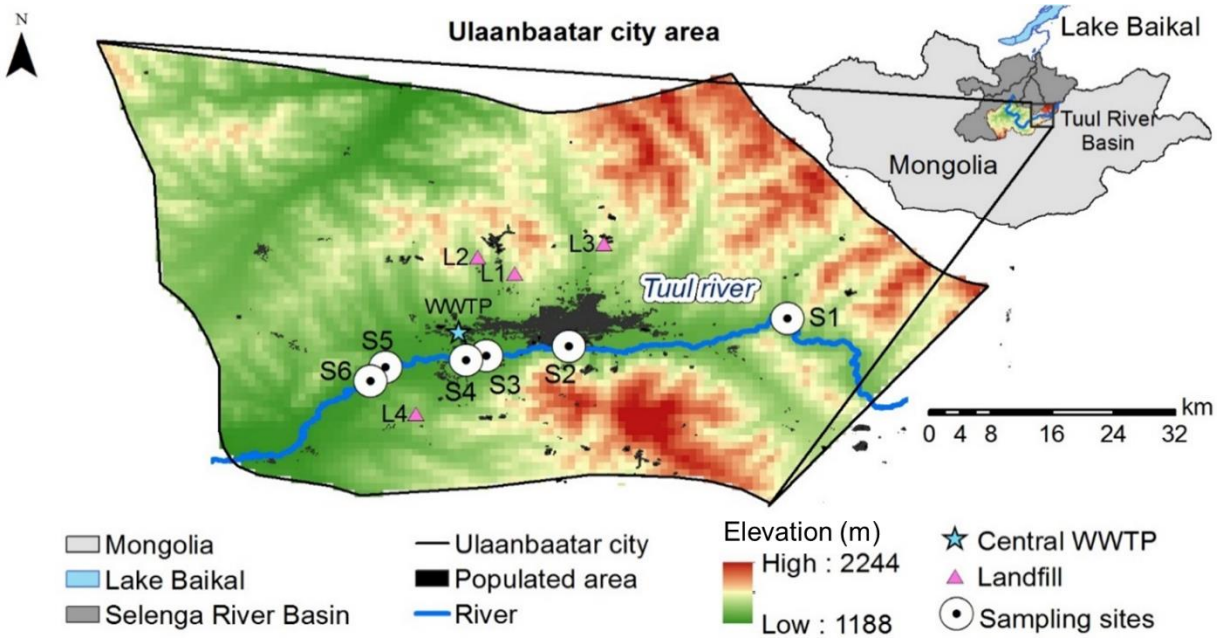

Fig. 4.1 Study sites along Tuul River in the Ulaanbaatar City area, Mongolia.

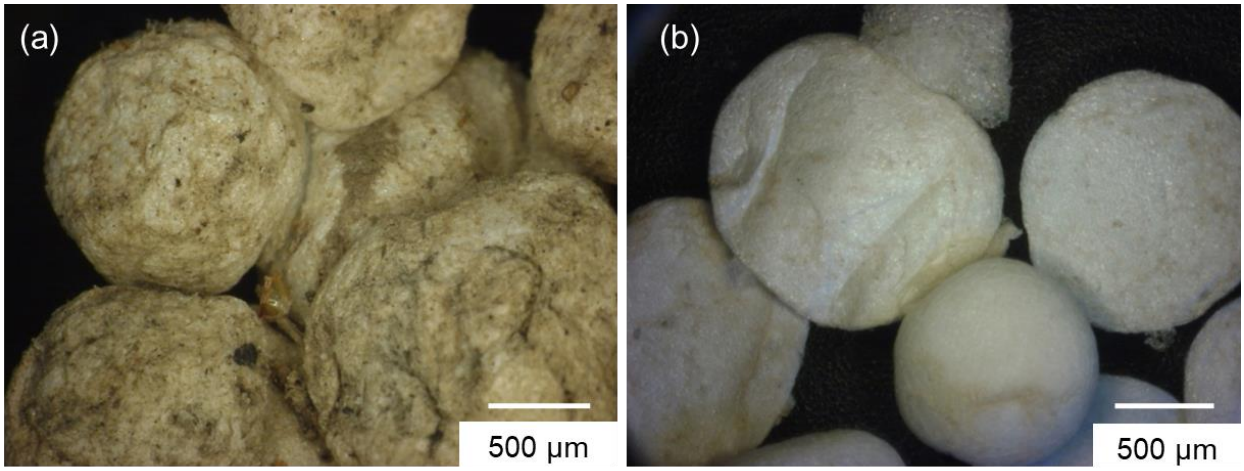

Fig. 4.2 An example of collected foamed plastics in research sites (a) collected from the field and (b) after organic matter digestion using peroxide with iron solution.

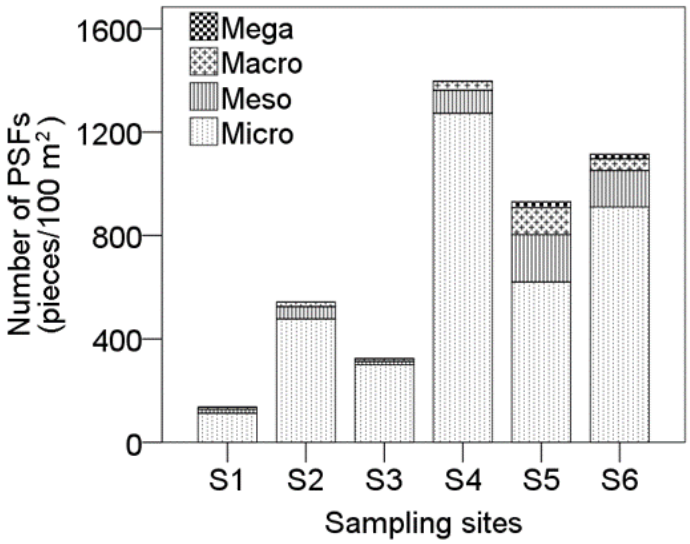

Fig. 4.3 Composition of sizes of PSF at each sampling site on the river shore of Tuul River.

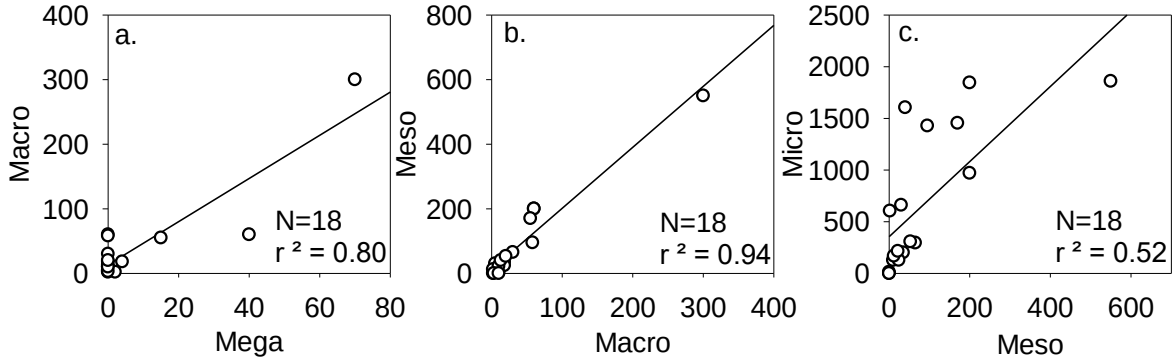

Fig. 4.4 Relationships between number of PSFs in size fractions distributed in the sampled area (100 m^2). a: mega- and macro-sized, b: macro- and meso-sized, and c: meso- and micro-sized fractions, respectively.

a.

b.

Fig. 4.5 Linear distribution of PSFs along the river shore (a); Distributed PSFs on the line, white dots and fragments shown as the piece of PSFs (b).

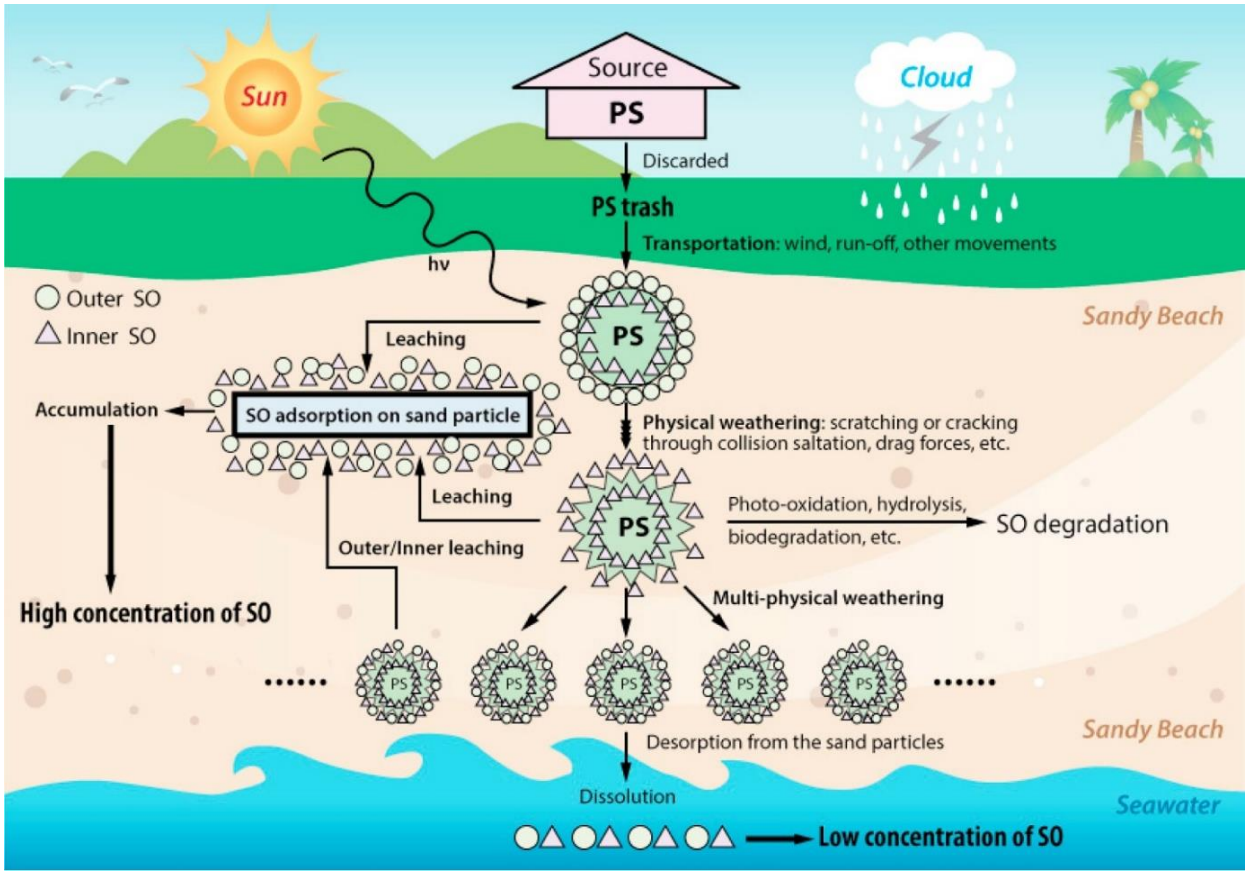

Fig. 4.6 Flow diagram of styrene oligomers leached from polystyrene pollution (Kwon et al., 2015). (PS: polystyrene; SO: styrene oligomer).

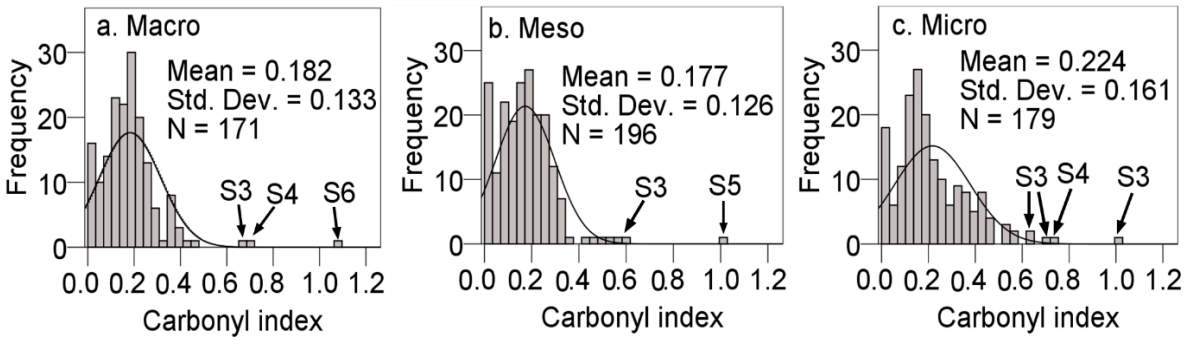

Fig. 4.7 Frequency of PSFs with a range of carbonyl index in (a) Macro-, (b) Meso-, and (c) Micro-sized PSF particles collected from the shore of the Tuul River. The relatively high value of carbonyl index (>0.6) was located at downstream from the city center. Solid curves were the normal distribution curve calculated by SPSS Statistic V22.

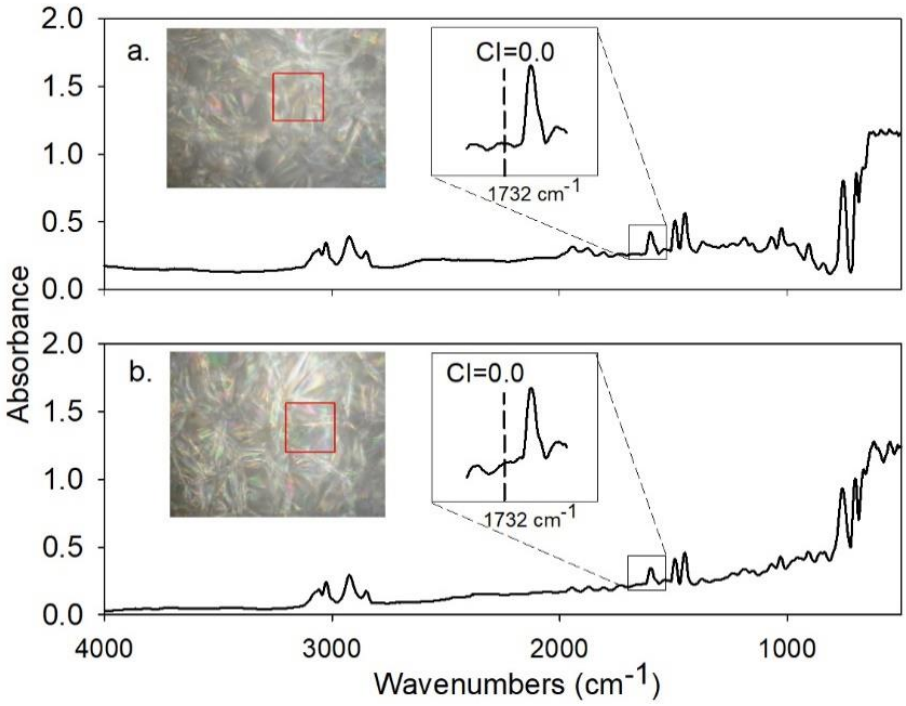

Fig. 4.8 IR spectra of reference the micro-PSF with microscopic images. a. Spectra of reference PSF before digestion; b. Spectra of reference PSF after digestion by the Fenton reagent.

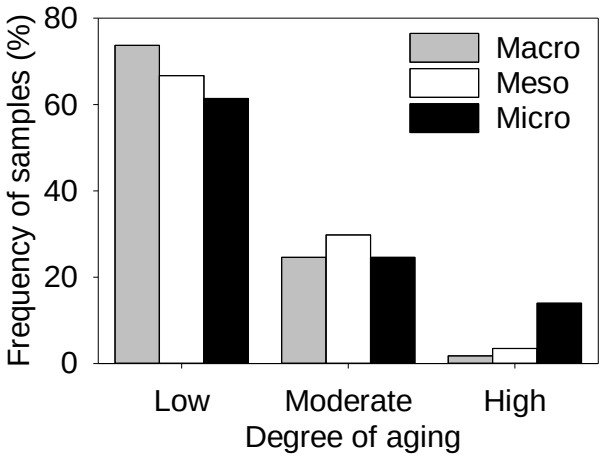

Fig. 4.9 Rate of frequency in degree of aging of macro-, meso-, and micro-sized PSFs categorized by carbonyl index.

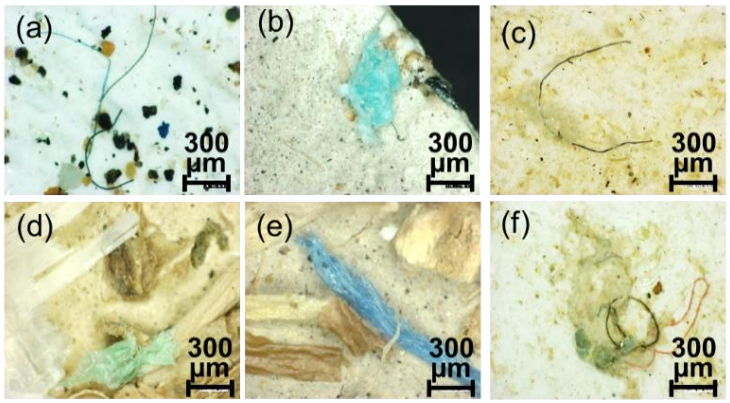

Fig. 4.10 Microscale pictures ((a)–(f)) of invisible adhered MPs. Invisible micro-fibers ((a), (c) and (f)) and micro-fragments ((b), (d), and (e)) adhered onto the surface of PSFs.

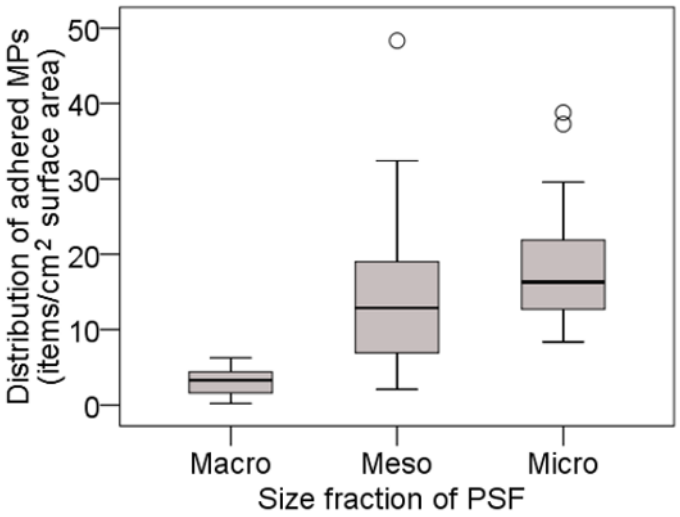

Fig. 4.11 Distribution of adhered MPs in a unit of surface area on macro-, meso-, and micro-sized PSFs.

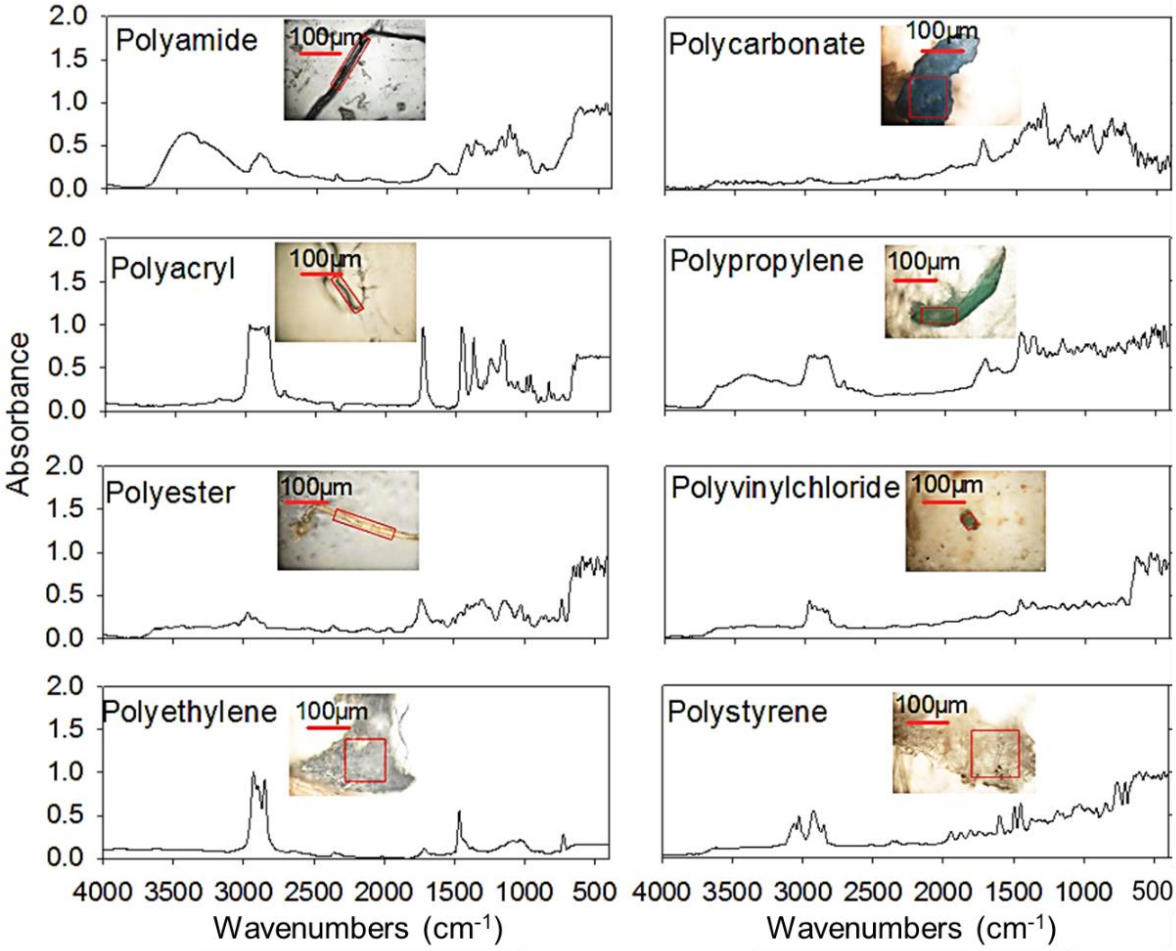

Fig. 4.12 IR spectra of adhered MPs obtained from PSF samples. The red square indicates the target area for the spectral analysis. The red line is a scale indicating a length of 100 µm.

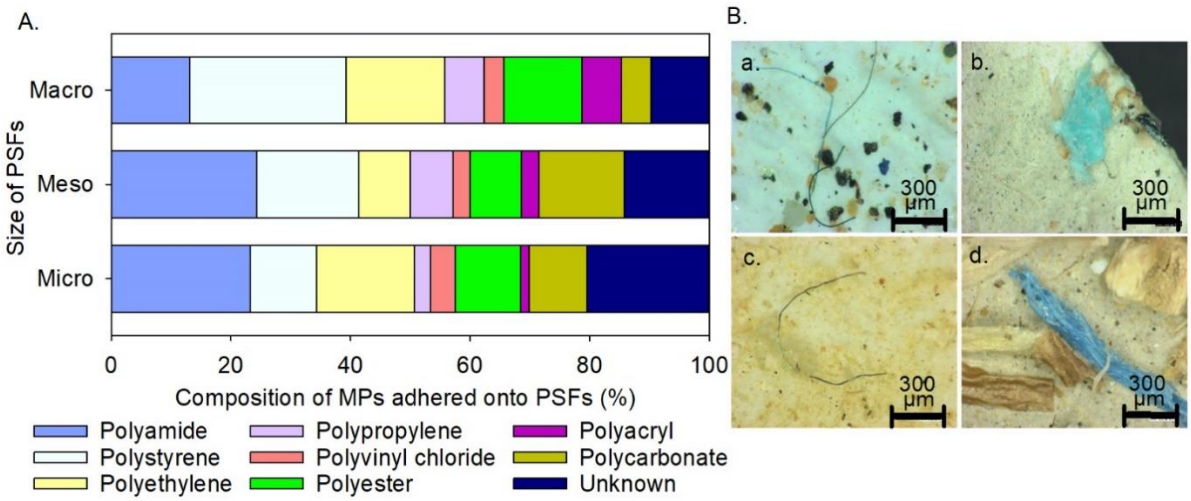

Fig. 4.13 Polymer composition (A) and microscale pictures (B) of invisible microplastics (MPs) adhered onto the surface of macro-, meso- and micro-sized PSFs. Invisible micro-fibers (a and c) and micro-fragments (b and d) adhered onto the surface of PSFs.

Table 4.1 Mean abundance of plastic debris on the Tuul River shore of Ulaanbaatar City area expressed by morphotypes and size fractions (Number of plastic pieces per 100 m^2 area ± SE).

	Foam	**Film**	**Fragment**	**Fiber**
Mega	7.3 ± 4.0	8.8 ± 1.6	5.1 ± 1.7	3.9 ± 1.8
Macro	36.6 ± 13.5	24.5 ± 3.3	11.3 ± 2.4	1.5 ± 0.7
Meso	81.5 ± 26.0	4.9 ± 1.2	6.6 ± 2.1	0.0
Micro	615.6 ± 157.3	4.4 ± 2.4	2.5 ± 0.5	0.3 ± 0.3

SE: Standard error

Table 4.2 Average number of adhered MPs in a piece of macro-, meso- and micro- sized PSF at each sampling site.

	Foam	Film	Fragment	Fiber	Total
Macro					
S1	26	24	27	28	105
S2	23	23	32	30	108
S3	16	15	15	18	64
S4	35	28	31	36	130
S5	22	22	36	31	111
S6	22	20	31	25	97
Meso					
S1	15	15	19	27	76
S2	30	34	40	37	141
S3	15	18	15	27	75
S4	13	9	16	14	52
S5	26	21	38	40	124
S6	24	26	35	28	113
Micro					
S1	2	2	2	2	7
S2	2	2	2	3	10
S3	2	2	2	2	8
S4	1	1	1	1	5
S5	9	9	12	12	42
S6	2	3	4	3	11

CHAPTER 5

Abundance of microplastics in sediments from the urban river in Mongolia

5.1 Introduction

Streams and rivers are considered as not only major pathways of plastic transportation to the open water systems but also reservoirs of plastics on the shore (Chapter 3 and 4) and in the bottom sediments (Anderson et al. 2016b). Importantly, recent studies estimated that benthic sediments have higher abundance of plastics compared to those in water columns (Hoellein et al., 2019). Microplastics (MPs) accumulate in river bottom sediments during their fluvial transportation (Blašković et al. 2018), causing a significant threat to habitats in the river ecosystem. As discussed in the Chapter 3 and 4, factors relating to climate and geomorphology of a river are the major controlling factors for not only plastic distribution in the environment but also plastic deposition into the bottom sediment and their interaction with suspended sediment particles in the stream ecosystem. Li et al., (2019a) have tested the formation of aggregates of micro and nano-sized plastic particles with suspended sediments and their settling properties under the laboratory condition (Fig. 5.1). From the Figure 5.1, humic substances, salinity and size of suspended sediments have influenced to deposition of plastics aggregates. Although environmental fate and deposition process of MPs in aquatic environment recently have been addressed in field and laboratory studies (Haave et al., 2019; Hoellein et al., 2019), very limited information is available from freshwater and estuarine ecosystems (Blettler et al., 2017; Horton et al., 2017b; Shruti et al., 2019).

On the other hand, MPs are currently impossible to remove from the aquatic systems due to their small sizes, chemical inertness and ubiquitous distribution (Brandon et al., 2016). Thus, it is important to understand the environmental fate of plastics that lead to accumulation and deposition of plastic particles. Due to changes in surface properties on plastic materials accompanied by their physical and chemical properties, gradual sedimentation of plastic pieces occurs in the river environment (Kowalski et al. 2016; Vermeiren et al. 2016). Associating with the distribution of plastics on the urban river shores, it is essential to evaluate the plastic particles in the river bottom sediment in order to figure

out overall processes relating to plastic abundance, distribution, deposition pattern of plastics in the inland river system.

As emphasized in the Chapter 3 and 4, the major plastic pollution of the study river is a consequence of existence of sewage plants with low capacity and improper waste management. Focusing on environmental plastic distribution in the river environment, previous chapters have provided comprehensive information relating to abundance, spatial distribution, degradation and fragmentation of plastics particularly polystyrene foam (PSF) and invisible MPs on the river shore. However, investigation of MPs in river sediments remains unexplored. Behavior and distribution of MPs in sediments make it possible to gain better understanding of the dynamics of plastics in the freshwater system. Given the limited knowledge on the MPs in urban river sediments, the current chapter is designed with the following objectives: (1) identification and quantification of MPs in river bottom sediments, (2) investigation of behavior of MPs in river sediments, and (3) comparison of the distribution of MPs among the world rivers.

5.2 Materials and methods

5.2.1 Geographical background of the study area and sample collection

The Tuul River basin located in the northern central part of Mongolia lies ranging between 48°30'N and 48°56'N, and from 104°48'E to 108°13'E (Fig. 5.2). The elevation declines from 2800 m at the south to 760 m at the north. Despite of the complex geological background (Choi et al., 2004), sedimentary rocks dominate in the major parts of the Tuul River watersheds (Dalai and Ishiga, 2013). Relatively fresh to weathered parent materials provides sediments consisting of clay, sand, and gravels to the flat basin occupied by the highly developed area of Ulaanbaatar City along the Tuul River valley (Dolgorsuren et al., 2012). Chestnut soil is the most common soil type which mainly distributed in the lower region affecting to alluvial deposition along the river system (Qi et al., 2008). The topsoil in the floodplain of the Tuul River has relatively coarse soil texture as loamy sand to sandy loam (Jarsjö et al., 2017). Soil erosion by water and wind is mainly associated with the intensive rainfall events (June – September) and aeolian processes (February – May) in the study area (Pietroń et al., 2017). The southern part of the river basin is covered with poorly vegetated grassland steppe, while forest-steppe vegetation has occupied in the northern part in the basin. The sparse vegetation in combination with grazing makes the landscape

susceptible to erosion and degradation. On the other hand, wind-driven fine particles may accumulate in snow and ice during winter and be transported into the stream network with spring meltwaters.

The study area was located in the Ulaanbaatar City along the Tuul River (Fig. 5.2). The Tuul River in this region connects with four main tributaries, which are Gachuurt, Uliastai, Selbe and Tolgoit streams located from east to west, and mainly flow in the middle of Ulaanbaatar City. The Gachuurt and Uliastai tributaries flow in the less populated areas in the eastern districts. However, domestic people enjoy camping and field recreations along Tuul River in Gachuurt village because of better water quality (Batbayar et al., 2017). The upper reach in the study area, where is approximately 40 km far from the city center, is comparatively less affected by residential areas. There were no industries along the Tuul River, but one of the largest organic farming agricultures was located in this area. The lower reach of the confluence with the Uliastai tributary, the Tuul River has been protected by government for the drinking water resource. The Selbe tributary run through the most populated area in the Ulaanbaatar City collecting all of drainage from residential and industrial areas. The watershed of the Tolgoit tributary has been occupied by residential areas of Mongolian Yurt as well as the concrete modern apartment areas. There was a social problem of waste management released from Mongolian Yurt (Byamba and Ishikawa, 2017).

Sediments collected from the Tuul River at the same time with collection of shore plastic samples (Chapter 4). The S1 plot was located at the upper stream of the Tuul River close to the confluence point with Gachuurt tributary (Fig. 5.2). The other plots were downstream of Tuul River from the confluence point with the Selbe to the downstream of WWTP. The S2, S3 and S4 were located between the confluence of the Selbe and WWTP. The S2 plot was the closest point from the mouth of Selbe, where was the prohibited area to enter. The area between Gachuurt to Selbe tributaries was skipped for sediment sampling because of less anthropogenic activities. The S5 and S6 sampling plots are the downstream plots of central WWTP where just after confluence of Tolgoit tributary.

5.2.2 Sample preparation

At each plot, triplicate samples were randomly collected to the depth of 5 cm using a stainless-steel shovel. All collected sediments were dried at room temperature for over two weeks. Then sediments were sieved into three fractions and separated by particle sizes. Gravels (>2 mm) were separated using a 2 mm sieve. The coarse sand fraction was collected

from the sediments previously passed through a 2 mm sieve using a 0.5 mm sieve. The sediment sample, which passed through a 0.5 mm sieve, was collected as the fine particle fraction. Microplastics in the three particle size fractions were used for further analyses. To remove natural organic materials, the size fractions of sediment samples were digested with the Fenton reagent (Masura et al. 2015). The digestion was conducted on a hot plate (at approximately 75°C) until no organic matter was visible in the mixture. Subsequently, the mixture was processed in an ultrasonic bath for several minutes to destroy aggregates of plastic and sediment particles. The extraction of MPs was then conducted using the density separation method (Ballent et al. 2016). Sodium polytungstate powder was added to the mixture to obtain a final density of 1.5 g cm^{-3} and stirred for 5 – 10 min using a magnetic stirrer. The mixture was allowed to settle for 20 min before being transferred to a centrifugation tube. Following centrifugation at 4000 rpm for 30 min, the supernatants were filtered through a 0.2 µm membrane filter (Millipore, Co Ltd.). A complete recovery of total MPs was achieved by carefully rinsing the filter and the filter holder with deionized water during the filtration process. Finally, the obtained samples were dried.

5.2.3 Plastic quantification and identification

The identification of plastic particles was conducted the same way as described in the Chapter 3 and 4. Morphotypes, sizes, and abundance of MPs were examined under the digital microscope with 100× magnification. Additionally, the observed MPs were categorized into four color categories (translucent, colored, white, and black) according to Crawford and Quinn (2017) and Shabaka et al. (2019). The average number was converted to the number of MP items in a theoretical area of the filter which collected whole of MPs from the applied sediment factions. In addition, the sizes of MPs were determined using a scale with a 50 µm resolution in the microscopic view. The size distribution of the MPs was classified based on their sizes with 100 µm intervals under the digital microscopic view.

Kernel density estimation (KDE) is commonly used to estimate the probability of density function of continuous random data (Rajagopalan et al. 1997). In this case, distribution of sizes of MPs is estimated employing the size data from the microscopic views. The KDE data were obtained utilizing the R program (R studio ver. 3.6.0) and using the following equation (*5.1*):

$$\widehat{f_n}(x) = \sum_{i=1}^{n} \frac{1}{nh_i} K\left(\frac{x - x_i}{h_i}\right) \qquad (5.1)$$

where K is the kernel function centered on the observation of i^{th} targeted plastics (x_i), h_i is the bandwidth of the size distribution or "scale" parameter of the kernel centered at x_i and n is the number of target MPs.

Infrared spectroscopic analysis was performed in the same way as described in Chapter 4 to identify the origin of MP materials.

5.3 Results and discussion

5.3.1 Occurrence and identification of MPs in sediment samples

The average number of MPs found in all sediment samples were 603 ± 251 items kg^{-1}. The highest number of MP was 998 items kg^{-1} recorded at S6, while the lowest was 312 items kg^{-1} at S2 (Fig. 5.3). The S6 plot was located at the downstream in the study area and it was exposed to daily discharge of wastewater from the central WWTP. Continuous deposition of plastic fragments from the city center through river flow enhances the number of plastics at S6. MPs sedimentation at plot S2 under the bridge could be avoided by herbaceous larch-birch forest grown along the river coast. Sources of MPs were diverse released from various land uses in the Tuul River watershed in Ulaanbaatar city. Less controlled waste management in the traditional Mongolian Yurts residential areas, recreational, agricultural, and industrial areas can release plastic wastes followed by their self-degradation into MPs (Batsaikhan et al., 2018).

In this study, the abundance of MPs at the plot S3 in fine sand fraction (<0.5 mm) were 455 ± 96 items kg^{-1}, revealing heavy load of MPs in the Tuul River after sewage discharge from the city center (Fig. 5.3). Despite of differences between sampling plots, significantly large number of MPs in the fine sand fraction were determined at all sampling plots compared to other sediment fractions (except for plot S6 in Fig. 5.3). On the other hand, MPs in >2 mm and 0.5 – 2 mm particle size sediment fractions were dominated at plot S6 probably due to large contribution of fibers supplied from central WWTP (Table 5.1). The diverse abundances of MPs at each sampling plot with different sediment fractions (Fig. 5.3) might be explained by environmental factors that affect sediment and plastic deposition (e.g., water

current, shoreline topography), and aging of MPs (e.g., degradation, fragmentation, physical and chemical changes, biofouling) (Ballent et al., 2016). Under the environmental condition, most plastic debris becomes brittle over time. These aged MPs may be covered with biofilms and eventually increase their specific density, causing their sedimentation. Additionally, during the travel of plastic debris, they interact with other substances in the aquatic environment and formed agglomeration with natural and anthropogenic products, resulting that increase in the density of agglomerates allows them to be precipitated into the sediment (Koelmans et al., 2017). These phenomena could affect deposition and distribution pattern of MPs in the river sediment.

Among the morphotypes, fiber-type MPs was dominant in the sediments, which was estimated to 35%. The other three morphotypes were similar distribution (Table 5.1). The fibers have been confirmed as a dominant plastic type in river and coastal sediments (Baptista et al., 2019). In the Chinese Wei river, the concentration of fibers was the highest compared to other types of plastics, which was accounting for 42.25 to 53.20% in the plastics in sediments (Ding et al., 2019). The predomination of fibers in marine bottom sediments of the Southern Baltic Sea have been reported by Graca et al., (2017). Therefore, the significant source of MP fibers might be laundry wastewater and they can be transported to river environments by wind (Haave et al., 2019). In fact, our sampling plots of S3 and S4 were located at downstream of the city sewage canal. Again, plots S5 and S6 were also positioned just after the wastewater effluent discharge, WWTP. Drainage of wastewater enhances contamination of micro-plastic fibers in river bottom sediments. Although visible sized PSF was dominant on the Tuul River shore (Chapters 3 and 4), it was not dominant plastics at the bottom sediment due to their light density as compared to fibers released from sewage and wastewater effluent. While the plastic depositions were confirmed at several sampling plots with geographically point-source locations, water stagnation observed in S3 and S6 plots can contribute to accumulation of MP fibers in the river sediments (Table 5.1). The similar spatial distribution patterns of MPs have been described in English Channel, UK by Browne et al., (2011). From the geographical viewpoint, proportionately more plastics were deposited downwind in habitats with slow-moving waters along the estuarine shoreline.

The morphotypes depending on colors (white, black, colored and translucent) were used to differentiate MPs. The predominant color group was white (36.6%) in >2 mm size fraction of sediment, while colored groups dominated in other size fractions of sediment with 35.4% (in 0.5 – 2 mm) and 32.0% (in <0.5 mm) (Fig. 5.4). The wide range of color indicated various sources of MPs, while white color was originated from dominance of PSF plastic debris in

the Tuul River sediment. In the collected MPs, the translucent group was mainly found in the films, whereas black and colored groups were derived from the fibers and fragments. Overall, MPs occurred in a variety of sizes, colors and types in the collected samples.

5.3.2 Size distribution of MPs and character of polymer

MPs occurred in a variety of sizes in the sediment fractions (Fig. 5.5). The size range of MPs ranged between 28.4 µm and 3,409.1 µm. The size of measured MPs mainly belonging to the range between 0 to 1000 µm (95.2%) in the sediments. The relatively large number of MPs were determined in sizes between 100 to 400 µm which occupied approximately 70% in each sediment fraction (Fig. 5.5 a-c). Other MP size categories such as below 1000 µm were captured less than 10%, whereas over 1,000 µm size categories were recorded less than 1% in the sediment samples. MPs with the size range between 0.5 and 0.99 mm occupied the largest proportion in precipitated MPs from Jiaozhou Bay, China (Zheng et al., 2019). While, Bergmann et al., (2017) reported that 99% of all particles were smaller than 0.15 mm in the Arctic deep-sea sediments. Another study in St. Lawrence River sediments revealed that most of (99.9%) microbeads below 2 mm diameter (Castañeda et al., 2014). These reported results and our study demonstrate that fragmentation leading to large number of smaller sized MPs occurred in the sediments.

In this study, various sizes and shapes of MPs were detected on the filter (Fig. 5.5 d-f). A number of MPs were regarded as degraded MPs which have minor cracks and rough surfaces. The dominant size distribution of MPs was 100 − 200 µm > 200 − 300 µm > 300 − 400 µm in >2 mm sediment fraction (Fig. 5.5a) while 100 − 200 µm > 300 − 400 µm > 200 − 300 µm was prevalent in 0.5 – 2 mm (Fig. 5.5b) and in 0.5 mm size fractions of sediments (Fig. 5.5c). The contributions of foams, films, fragments and fibers were distinctly observed in each sampling plot (Fig. 5.6). The predominated MP size was 100 − 200 µm in foams (54.6%), films (38.7%), and fragments (56.0%), and 300 − 400 µm size in fibers (20.1%). Observed MPs in the Tuul River sediment are secondary and primary MPs. Primary MPs were directly released from sewage and wastewater effluent from WWTPs, whereas secondary MPs were generated through breakdown of primary plastic litter along the river (Andrady, 2017).

Among the size distribution of foams, films and fragments in each sampling plot, most of MP particles were found less than 500 µm sizes, however, a number of MP items were reached to 3,000 µm (Fig. 5.6). The dominance of smaller sized foams, films and fragments

emphasize that various types of plastic debris accumulated along the river and breakdown into smaller size particles under the environmental condition. However, micro-fibers were identified with wide range of size distribution in each sampling plot (Fig. 5.6d). Considering a broad range of size in fibers at each sampling plot, the fibers originated from domestic washing machines and discharge of wastewater from WWTPs to the river (Napper and Thompson, 2016). Moreover, most of traditional Mongolian Yurts districts have not developed sewage water system with sufficient capacity, resulting that discharged water leads to soil and groundwater pollution by waste plastics (Itoh et al., 2011). Accumulation of synthetic fibers is one of the significant reasons of contaminated soils and groundwater in Tuul River system through infiltration and surface runoff. Significant accumulation of micro-fibers with a wide size range in S1 and S4 plots (Fig. 5.6) might be explained by inappropriate release of sewage water from domestic washing machines to the river watershed from the nearest Mongolian Yurts residential areas. Although wastewater discharge at S5 plot was the most relevant source of micro-fibers with broad size ranges, fine MP particles were deposited at the plot S6 after the short distance movement with river flow (Fig. 5.6). Plastic fragments were derived from a various type of discarded plastics such as food packages, polyethylene terephthalate bottle, plastic carry bag, construction materials in the river system (Rodrigues et al., 2018a).

The collected MPs in sediments were identified as polyester, polyethylene, polystyrene, acrylonitrile butadiene styrene, polyvinylchloride and polyamide from FTIR spectra (Fig. 5.7). The most common plastic materials made from polyethylene, polyvinylchloride and polystyrene in the world plastic market (PlasticsEurope, 2017) have been dominantly found in the Tuul River sediments. Most of fibers contained polyester and polyamide polymers which are also one of the widespread plastic fibers in the aquatic environments (Kanhai et al., 2017). Plastic material density and its surface status influenced retention of MPs in sediments. Floating polyethylene plastics initially cannot sink due to its low density (<1 g cm^{-3}), while aged polyethylene plastics lose buoyancy because of biofouling and changes in surface status mainly by photo-oxidation (Kaiser et al., 2017). The non-buoyant polymers (polyamide, polystyrene, polyvinyl chloride and acrylonitrile butadiene styrene) were also widely detected from the IR spectra (Fig. 5.7). Overall, our results emphasize that a large number of detected MPs have been influenced by environmental and anthropogenic factors which leads alteration of surface property of plastic materials and this reformed surface status affect the distribution pattern of MPs in the aquatic environment.

5.3.3 Comparison of MPs content with worldwide studies of environmental plastics

A several studies have been reported MPs in the river sediment as shown in Table 5.2. In comparison with other research reports in the river system, the abundance of MPs in Tuul River sediments was one or two orders of magnitude lower than St. Lawrence River and Atoyac River (Table 5.2). However, comparable amounts of MPs were determined in River Thames (UK), Wei River (China) and rivers in Shanghai (China) (Ding et al., 2019; Horton et al., 2017a; Peng et al., 2018). Most of MP particles could be originated from distinct sources including wastewater and sewage effluents from urbanized areas and discharged to the Tuul River. However, several studies highlighted MPs from highly industrialized areas (Alam et al., 2019; Shruti et al., 2019). Moreover, most of studies used different sampling methods and analyzing techniques for identification of MPs in the sediments (Eerkes-Medrano et al., 2015). The differences in methodological approaches and pollution sources could be significant reasons of various abundances of MPs in the river sediment.

The differences in results also suggest that local parameters such as wind, precipitation, air temperature and river flow affect the MPs transportation and accumulation in the river environment (Nel et al., 2019; Piperagkas et al., 2019). Since, spring season experienced snow-melting and strong wind events in Mongolia, it could increase flow rate and lead to mobilize sediments and previously settled plastic particles. On the other hand, the degradation of larger plastics from terrestrial sources such as landfill or litter must take into account for further distribution of MP particles in the river environment. Degraded plastics or aged MPs significantly ingested by various aquatic organisms (Vroom et al., 2017), leading to serious ecological risks. On the other hand, smaller sized MPs can enter to the higher to lower trophic system, which arise the further adverse impacts in the aquatic ecosystem. Recently, the ingestion of MPs by freshwater fish has been reported in the river environments (McGoran et al., 2017). From the reported results, 45% of sunfish have exposed MPs in Brazos River Basin, USA (Peters and Bratton, 2016), while 11 different species fish from Río de la Plata estuary in Argentina verified MPs in their gut contents (Pazos et al., 2017). The toxic effect of MPs is mainly intestinal damage and oxidative stress for aquatic biota (Lei et al., 2018). In case of Mongolia, there is no record of MP exposure in aquatic organisms. However, high abundance of MP has been confirmed in the Selenga River system as introduced in chapter 3. High number of plastic distribution in the largest freshwater lake Hovsgol has also reported (Free et al., 2014), resulting in increase of the probability of exposure to MPs for some species of fish. For these reasons, comprehensive

understanding of MP behavior in river environment and exposure scenarios are required by increasing attention because of growing concerns of ecological consequences due to MP pollution. Therefore, more studies that cover the spatial and temporal distribution patterns and risk assessment of MPs are currently required to comprehend the interaction of these pollutants with other natural or anthropogenic substances in the aquatic environment.

5.4 Conclusion

In this study, the abundance, distribution pattern and behavior of MPs in the urban river sediment of Mongolia have been investigated. Although sediment samples from Tuul River contained various size and shape of MPs, the dominance of smaller size (100 − 200 μm) and fibrous MPs illustrate that the studied urban river has received heavy load of released MP particles and fragmented MP debris. Moreover, most of the plastic debris appears to be smeared with organic substances such as humic substances or algae, resulting in their deposition as sediments. It is noteworthy that fine particle sediments (<0.5 mm) have a great potential to collect MPs in the river environment.

Regarding to environmental fate and behavior, hydrodynamic forces (turbulence, stratification and plume fronts) influence on plastic items to follow circulation, dispersion, suspension, and settling pathways, which are the reason for hotspots distribution in the downstream of the river. In addition, point and non-points sources of pollution and polymer property have been led to unexpected accumulation patterns of MPs in the fluvial ecosystem. Furthermore, present study, which highlighted the behavior and distribution of MPs in the river bottom sediment, illustrates the novel aspects of widespread aquatic plastic pollution in terrestrial inland river system. In future research work, MPs aggregation and their formation mechanisms, and aggerates ingestion by terrestrial aquatic biota are necessary to investigate in order to understand adverse effects of MPs on wildlife and the ecosystem.

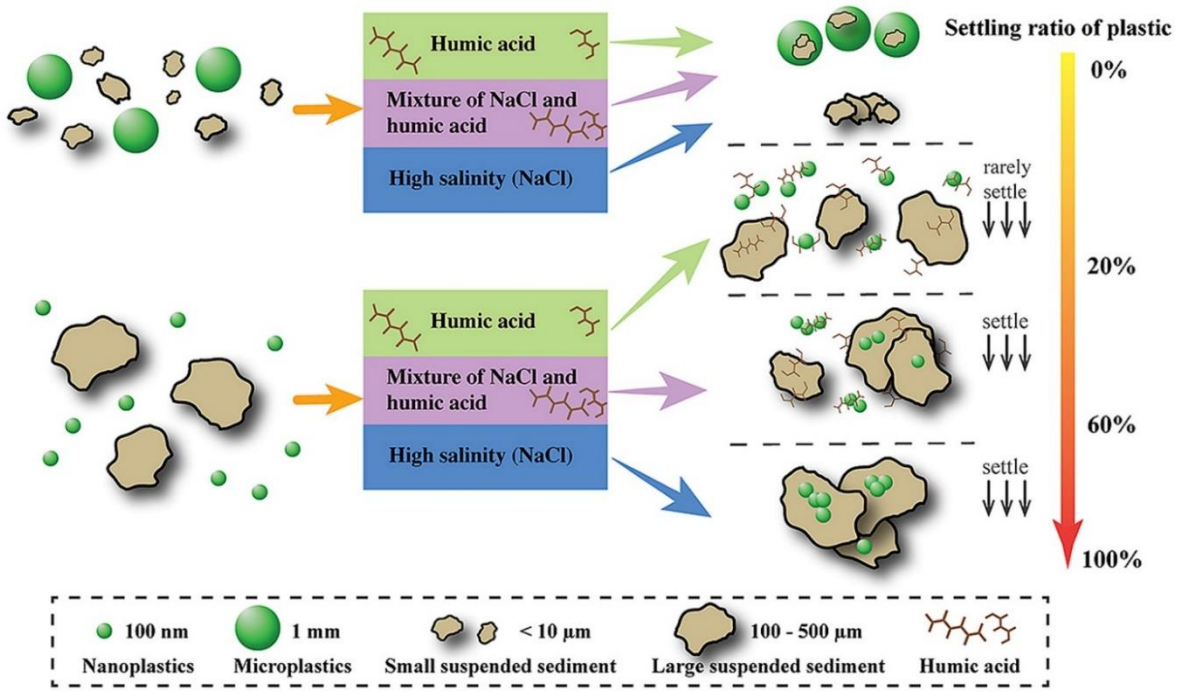

Fig. 5.1 Formation of aggregates of micro and nano-sized plastic particles with suspended sediments and their settling properties under the laboratory condition. The settling ratio of nanoplastics/microplastics as a function of time was calculated by Li et al., (2019b). (See more details in Li et al., (2019b)).

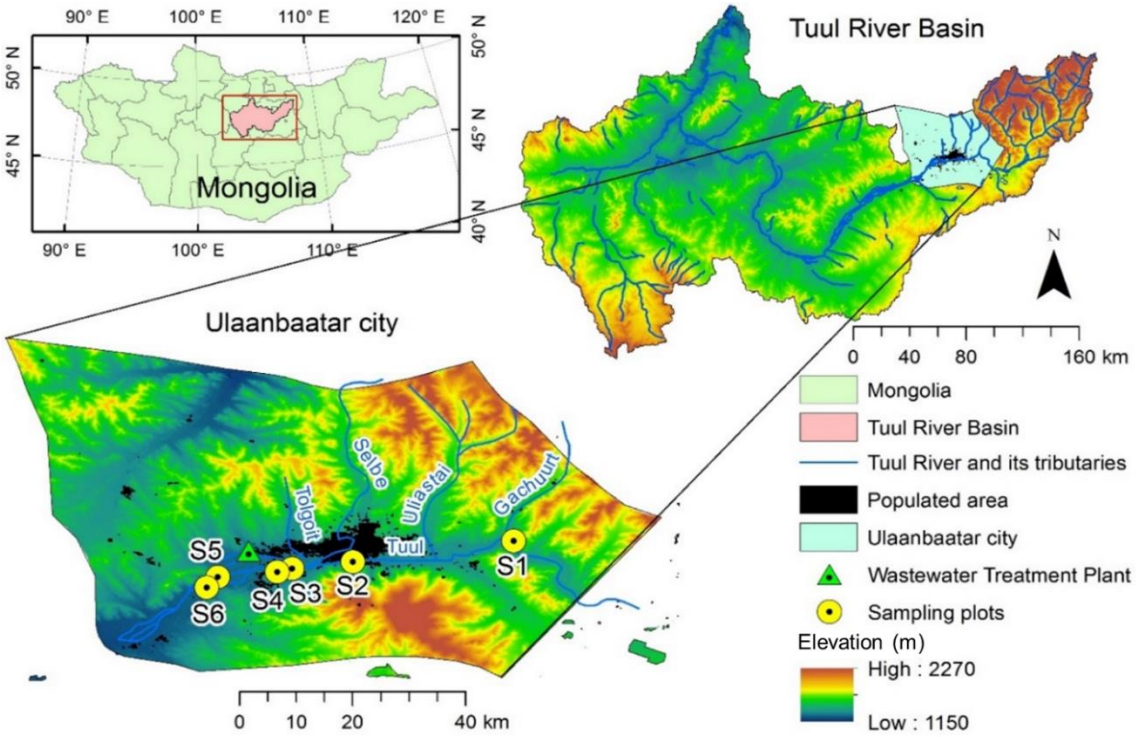

Fig. 5.2 Study area and research plots.

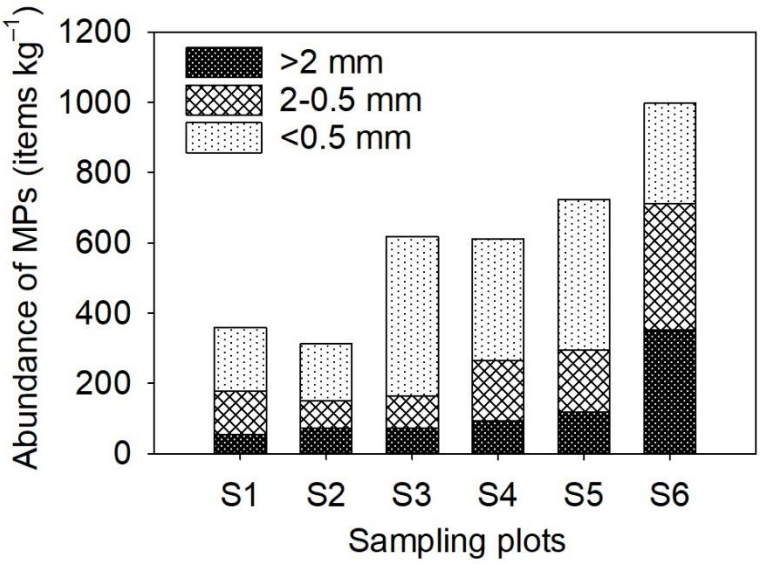

Fig. 5.3 Abundance of MPs in the three particle size sediment fractions at each sampling plot.

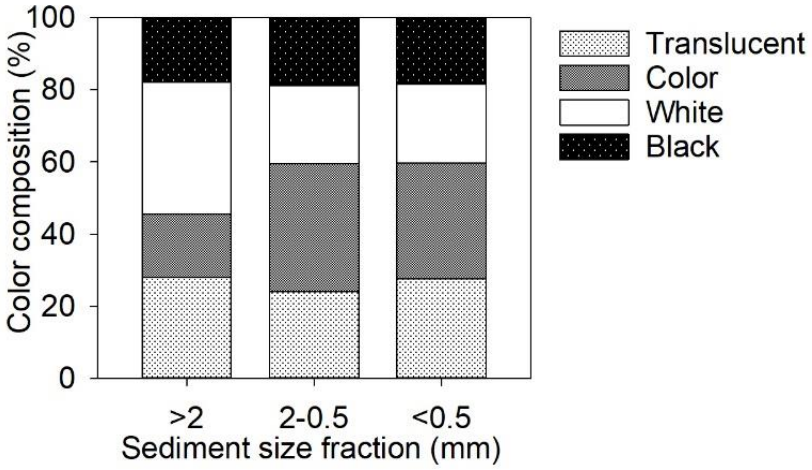

Fig. 5.4 Color composition of MPs extracted from sediment samples.

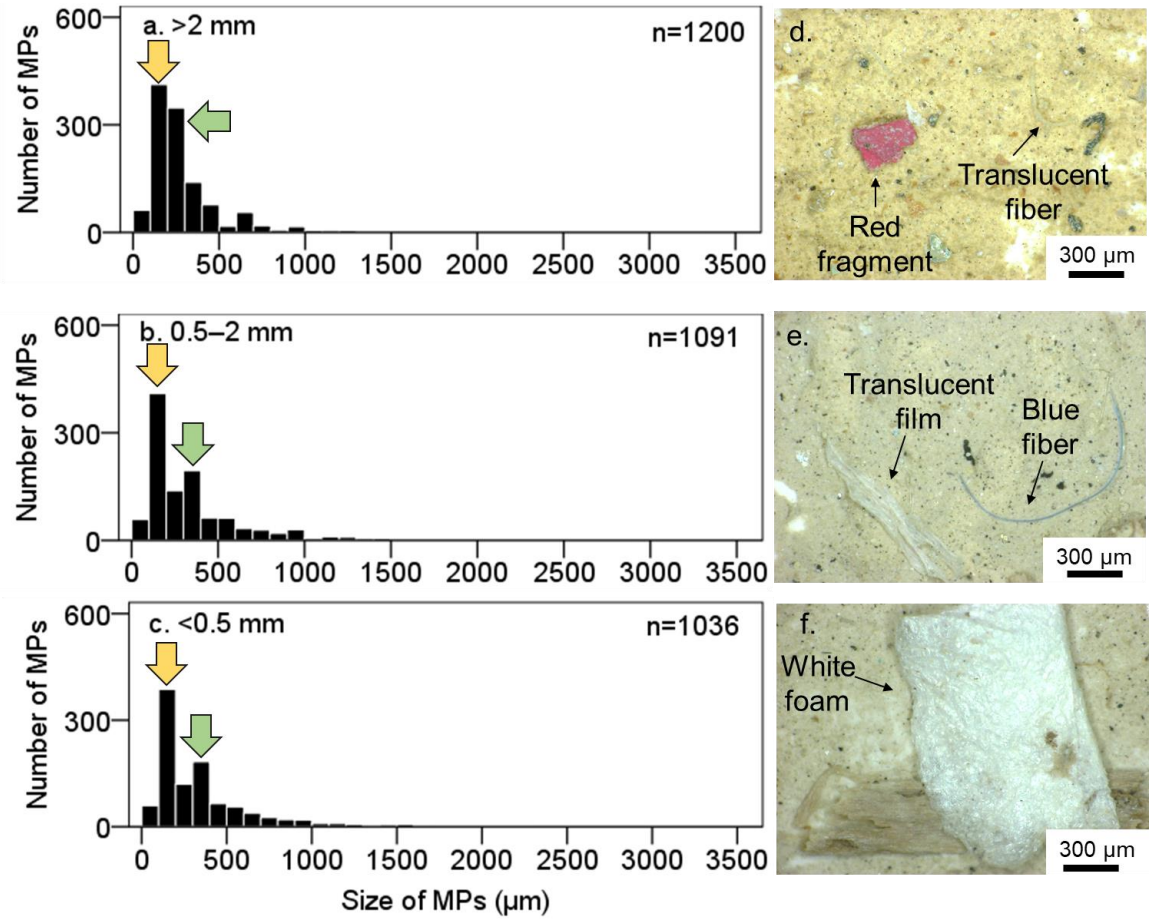

Fig. 5.5 Size distribution of MPs in (a) >2 mm, (b) 0.5 – 2 mm; and (c) <0.5 mm size fractions of sediment samples and microscopic images of morphotypes of plastics (d-f). Arrows in (a) indicate predominant size composition of MPs. Yellow arrow indicates the highest, and green arrow indicates the second highest size in the distribution of MPs in the sediment fractions.

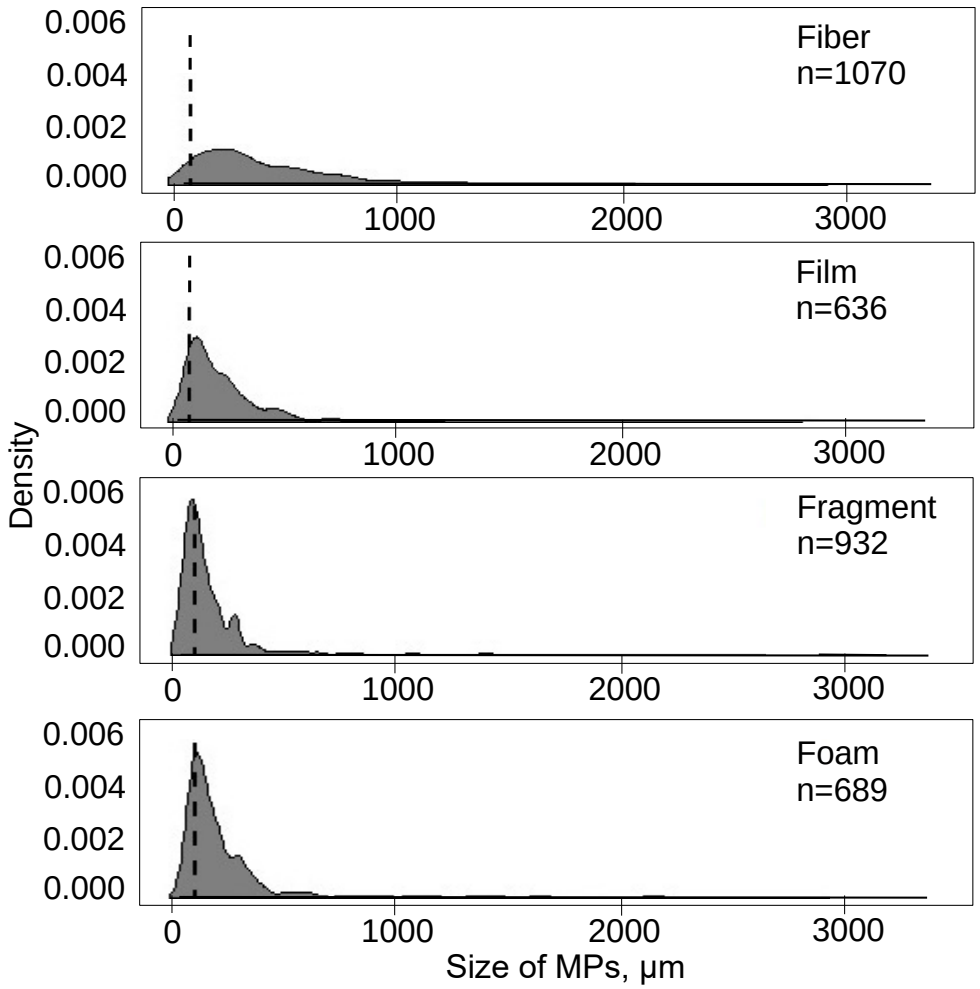

Fig. 5.6 Distribution of Kernel density calculated by R program with the dependency of particle sizes morphotypes of MPs in the river bottom sediments. The higher the Kernel density, apparent frequency of abundance is higher. The dashed line indicates 150 µm which was the highest density observed in the Foam and Fragment type of plastics.

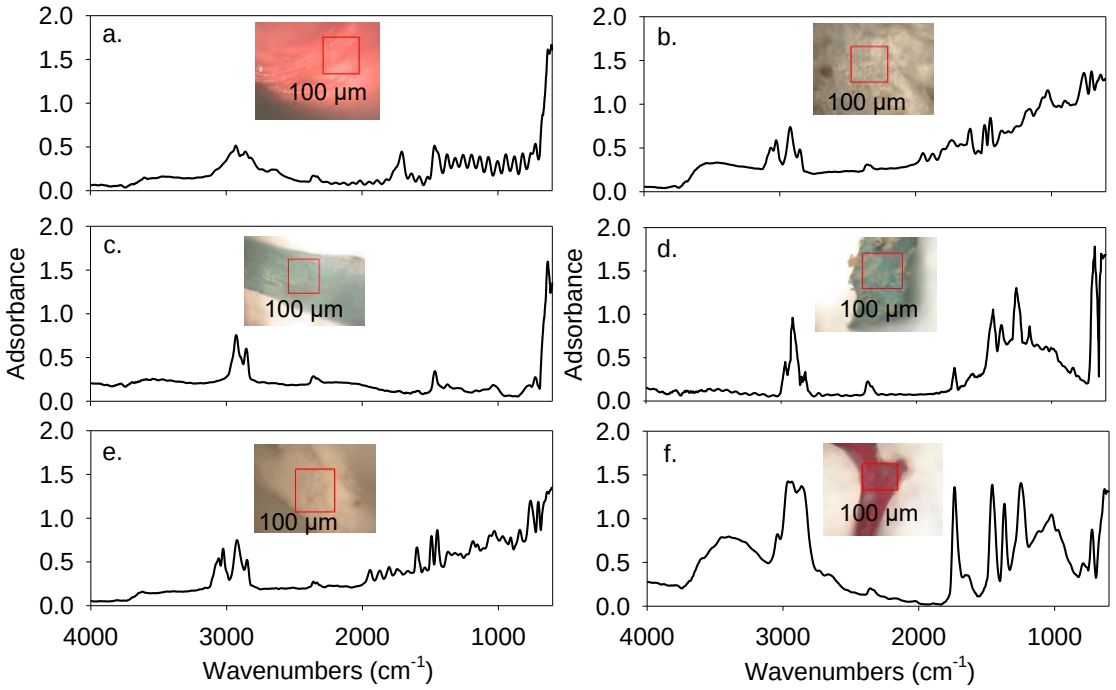

Fig. 5.7 FTIR spectra of MPs extracted from sediments. The spectra identified as a. polyester, b. acrylonitrile butadiene styrene, c. polyethylene, d. polyvinylchloride, e. polystyrene, and f. polyamide polymers using Aldrich library of FTIR spectra.

Table 5.1 Average concentration of MPs by morphotypes in the studied plots. (n=3; items kg^{-1} ± SD).

Sampling plots	Foam	Film	Fragment	Fiber	Total
S1	84±13	72±9	74±18	129±24	358±41
S2[†]	78	68	84	82	312
S3	125±28	96±15	153±57	245±51	619±126
S4	132±9	133±16	160±4	183±19	609±30
S5	142±19	162±33	185±47	234±24	723±70
S6	171±15	160±5	199±17	468±380	998±373

[†]without replicates

SD: standard deviation

Table 5.2 A comparison of abundance and size distribution of MPs in river sediments across the worldwide sampling locations.

Country	Location	Abundance of MPs	Dominant size classes	References
UK	River Thames	185 ± 42 items kg^{-1}	1 mm – 2 mm size was higher than 2 – 4 mm size	Horton et al. (2017)
China	Rivers in Shanghai	802 ± 594 items kg^{-1}	<100 mm - 31.19%, 100 – 500 mm - 62.15%:	Peng et al. (2018)
Indonesia	Ciwalengke River	30.3 ± 15.9 items kg^{-1}	1000 – 2000 µm size was higher than 50 – 100 µm	Alam et al. (2019)
China	Wei River	360 - 1320 items kg^{-1}	<0.5 mm is 40.8% to 68.8%	Ding et al. (2019)
Mexico	Atoyac River basin	4500 ± 702.23 items kg^{-1}	-	Shruti et al. (2019)
Mongolia	Tuul river	603 ± 251 items kg^{-1}	100 – 200 µm size	This study

CHAPTER 6

Conclusion

6.1 Distribution and characteristics of plastic debris in the river system in Mongolia

Environmental plastics have various properties processed by degradation, fragmentation, formation of biofilm and interaction with other pollutants according to their residential time in the environment. Some of their characteristics including size, morphotype, color and origin regarding to the behavior of plastics in the urban river environment have been discussed in this thesis.

Current study aimed to provide fundamental knowledge, distribution and behavior of plastic debris in the river environment, particularly urban river environment with respect to their physical and chemical properties (Chapter 1). The research was initiated with the purpose of establishment of appropriate sampling and methodological techniques to find, quantify and determine plastics in the river shore and sediments by reviewing broadly employed methodological approaches (Chapter 2). Although various methodological approaches have been applied to the plastics in the marine environment, those in river shores and sediments were not well established. Therefore, methodologies for river environments were highlighted in order to collect various sized plastics and analyze their characteristics based on differences between hydrological backgrounds. To apply the established experimental procedures to the determination of plastic samples in the Mongolian river systems, the characteristics including sizes, morphotypes, colors and origins regarding to the behavior and distribution of plastics in the river environment were pointed out in Chapter 3-5.

Visible plastic samples collected from the river shores of Selenga River system by hand-sorting technique to identify their abundance and distribution (Chapter 3). Spatial distribution of plastics and their fragmentation processes demonstrated the abundance of various sized plastics (mega: >100 mm; macro: 20 – 100 mm; meso: 5 – 20 mm; and micro: <5 mm) with different morphotypes (foams, films, fibers and fragments) in the Selenga River system. The 2 to 506 pieces per $100m^2$ area of plastic abundances at the river shores reflected multiple sources of plastics at the river watersheds relating to concentrated population (~80%), industrial (~70% of total industrial production) and agricultural (~60% of total agricultural production) activities and inappropriate waste managements in the Selenga

River system. Spatial distribution of observed plastics indicated that a large-scale distribution could not show any specific trend of plastic accumulation. Therefore, occurrence of plastic pieces at the remote locations revealed that the nearest urbanized areas are the major source of released plastics in the environment, which were mega- and macro- sized plastics. The fragmentation of plastics can be distinguished by characteristics of plastic materials during their travel by river transport. The distribution and positive significant correlations between size fractions of foams ($p < 0.01$) and films ($p < 0.001$) showed the evidence of on-site fragmentation in the river environment. Polystyrene foam (PSF) plastics were characteristic on the shores of Selenga River system based on frequent consumption and low recycling rate of generated plastic waste.

To investigate distribution of plastics particularly PSF debris, plastic samples collected from the shores of Tuul River (Chapter 4). The PSF samples were evaluated in terms of their chemical structural properties (surface oxidation), and the adhered MPs which were extracted from the surface of PSFs categorized into four morphotypes and identified chemical structure. Since large amounts of various sized (PSF) plastics were recorded on the river shores of Selenga River system at the high-populated areas, further concerns regarding to their transportation and mean residential time were highlighted in the urban river environment (Chapter 4). The values of carbonyl index (0.0 – 1.09) demonstrated that majority of determined macro-, meso- and micro- sized PSFs changed their surface chemical structure through photodegradation under the UV light. Although several PSF samples especially micro-sized PSFs had high value of carbonyl index (>0.6) regarding to their longer residential time, most of PSFs with less degree of aging (carbonyl index: 0.0 – 0.2) indicated lower residential time at the river environment. Largely occupied PSFs on the shores of Tuul River considered as a carrier of other invisible plastic particles through their aggregate formation. Given to the diverse residential time in the environment, physical and chemical changes of PSFs and coverage of biofilms (biofouling) altered the aggregation potential. Furthermore, the characteristics of PSFs and adhered microplastics (MPs) were important indicators to identify the formation of aggregates, whereas mechanism of interaction was not clear due to diverse origins and distributions in the composition. Nevertheless, in order to understand behavior of plastics in the aquatic environment, it is necessary to take the plastic-plastic aggregates into account as a significant factor of plastic fate in the environment.

Regarding to the ubiquitous distribution of plastics in the river environment (Chapter 3), river bottom sediments tended to be a reservoir of environmental plastics (Chapter 5).

Sediments collected from the Tuul River and MPs extracted from the sediment size fractions (5 – 2 mm, 2 – 0.5 mm, <0.5 mm) in order to determine the morphotypes, sizes, and abundance of MPs in the river bottom sediment. The size range of MPs ranged between 28.4 µm and 3,409.1 µm. Microplastics with smaller sizes (100 to 400 µm sizes: 70% of total size distribution) and dominance of fibrous types (35% of total morphotypes) reflected the point sources of plastic debris into the urban river environment. The low specific densities of MPs are positively buoyant and thus likely spend a long time at the surface (or in the water column), where they can potentially be transported over long distances. The high proportion of MPs in the river sediments supports a possible mechanism to explain the missing plastic fraction in the surface of water. As shown highlighted in the Chapter 5, the study found not only non-buoyant plastics (polyamide, polystyrene, polyvinyl chloride and acrylonitrile butadiene styrene), but also buoyant plastics (polyethylene) which were preserved at the river bottom sediments. The settling mechanisms of plastics into the river bottom sediments were difficult to speculate in the natural system. However, the accumulated buoyant plastic particles in the river bottom sediments are the evidences for the increase of specific density of plastics as a result of changes in surface properties by biofouling of plastics. The abundance of MPs (312 to 998 items kg^{-1}) found in sediments from urban river network demonstrates an important sink for MPs in riverine sediments.

Diversity in abundance and characteristics of the environmental plastics in the Selenga River system and Tuul River system was underlined for the multiple pathways of plastic debris in the river catchment including the urbanized areas. Accumulation of plastic debris in the river environment might be brought by inappropriate management of municipal solid waste (MSW) through its landfill process without proper planning in Mongolia and uncontrolled release of plastic waste through unintentional anthropogenic activities. Collectively, the results of the plastic distribution, interaction and accumulation provide a fundamental insights into the plastic migration and preservation in the river environment from the viewpoint of plastic pollution.

6.2 Fate and behavior of plastics in the urban river environment of Mongolia

Current thesis emphasized the ubiquitous distribution and various properties of plastic debris in the inland river system. After a scientific and public attentions addressed to the environmental plastics, recovered plastics from the river environment has been reported through clean-up activities (Chapter 1). However, majority of released plastics are

unrecoverable due to their small size and unforeseeable distribution by ingestion or sink to bottom sediments in the environment. According to the predominance of MPs in any aquatic compartment, both of recoverable and unrecoverable plastic debris (Fig. 6.1) can be considered as “lost-plastics” or “missing-plastics”. A large proportion of the plastic fragments are lost from the surface due to hydrodynamic processes (Eriksen et al., 2014). As a consequence of complex natural phenomena, it is impossible to fully speculate the fate and distribution pathway of environmental plastics not only in inland river system but also in coastal and marine environments (Dikareva and Simon, 2019; Yan et al., 2019). However, the linear relationships between size fractions of plastics in Selenga River system and the abundance of various sized PSFs at the Tuul River system illustrate the plastic fragmentation which is significant evidence of the “lost-plastics” through the formation of invisible micro and nano-plastics. Furthermore, the abundance of MPs in the Tuul River sediments provided important sink for plastics. These results provide further important information for the understandings of possible mechanisms as the fate of plastics in the aquatic environment.

Figure 6.1 shows the fate of environmental plastics. After plastic released into the environment, fragmentation, formation of aggregates (or interaction with other particles and pollutants) and deposition processes are the key factors to demonstrate the fate of environmental plastics. The fragmentation process decreases the size and increases the number of items in the aquatic environment. The deposition process is based on hydrodynamic processes of river. Based on specific density of plastics, settled plastics, which is usually non-buoyant plastics, can be occurred after their release into the aquatic environment. Therefore, buoyant plastics also can be preserved into the bottom sediment of river after the increase of specific density through the formation of aggregates. Overgrowth of micro- and macro-organisms causes an increase in specific density and sinking of plastic particles (Chubarenko et al., 2016). According to the diverse environmental processes, homo-aggregates consisting of plastic-plastic interactions or hetero-aggregates by coupling between plastics and suspended sediments can be major aggregate plastics, affecting their fate and distribution in the aquatic environment (Fig. 6.1). The homo- and hetero-aggregates are considered as the significant characteristics of environmental plastics.

As highlighted in Chapter 4, formation of plastic-plastic aggregates (homo-aggregates) is the new insight of current thesis and the interaction between plastics in the environment could not be limited by only PSF and invisible MPs. Other morphotypes and polymeric structures of plastics (e.g., polyethylene films, polypropylene fragments, resin pellets) largely distributed in the aquatic environments. Based on hydrophobic interactions between

the plastics, it is possible to find various plastic aggregates from the aquatic systems. Moreover, plastic-suspended sediment/sediment particles (hetero-aggregates) are also another aggregation status of plastics. Some studies demonstrated that aggregates of micro- and nano-plastics with suspended sediments in the laboratory condition in order to estimate rate of plastic settling using theoretical approaches (Li et al., 2019b). As shown in Chapter 5, the abundance of plastics reflected by the settlement of plastics from the flotsam into the river bottom sediments. Deposited plastics largely found at the <0.5 mm sediment fraction which indicates that MPs likely interact with fine-sediments particles.

In addition, hydrodynamic processes also influence the fate of environmental plastics. A turbulent condition would influence to the (re)suspension of plastics at the river environment. Considering the various particle sizes of plastics at the river shores and sediments in Tuul River system, dynamics of plastics at the river environment would characterize different suspension rate of smaller plastic debris in the water column and limit their sinking. Fragmentation and degradation of plastic debris within the turbulent river condition can also be responsible for size-driven accumulation of plastic pieces within the turbulent zone. After the high flow season, plastic pieces accumulate on the shores based on differences in sizes and morphotypes with low water level. As mentioned in the Chapter 4, the linear distribution of PSFs along the river shores could be an evidence of this hypothesis. However, more research is needed to understand the interaction between plastic-plastic aggregates and effects from hydrodynamic processes in the river environment.

Nevertheless, abundance of plastics in the Tuul River environment is the evidence for the extensive consumption of plastic products as a modern technological outcome in Ulaanbaatar City. Wide range of applications of plastic products are broadly used in the high populated areas. While inappropriate waste management and uncontrolled release of plastics into the environment (discussed in the Chapter 3) are the major reasons of plastic density in the river environment and their movement by river transport. The open water system, in this case Lake Baikal can be considered as the final destination of plastic debris. In this thesis, a footprint of anthropogenic products especially plastics has been identified in the river environment regarding their fate and behavior. The results of current study support the terms of “Plastisphere” because of ubiquitous distribution, unrecoverable release into the environment and danger to the living organisms. The urban river system considered as mediator of plastic debris and it is difficult to figure out important processes of plastics through this mediator. However, current study presents for the first-time plastic-plastic (homo-) aggregate in the natural river system. Furthermore, focusing on the hetero-

aggregates, deposited plastics have been addressed and reported by a potential interaction between plastics and fine-particle sediment fractions. The emphasized aggregates, properties of aggregated plastics and the formation mechanisms are the novelty of this thesis in order to understand the fate and behavior of environmental plastics.

6.3 Environmental plastics - research direction and knowledge gaps

Understanding sources, abundance and composition of plastics present in the environment is a huge challenge due to the large amount of plastic materials manufactured for annual societal uses, some of which is released to the environment. Although freshwater in terrestrial environments are recognized as sources of their transportation of plastics to the oceans, there is still lack of knowledges about these environmental compartments. Based on diverse consequences of plastic debris in the aquatic ecosystem, future research direction of environmental plastics is focused on dynamics of plastic particles in the environment and evaluating their behavior in order to fill gaps between knowledges of environmental plastics in the ecosystem. Following directions are considered to be the major research direction of environmental plastics.

1) Development of standardized methodology to monitor plastics particles in aquatic environments
2) Quantification of presence, abundance and distribution of plastics in the environment
3) Understanding of degradation status, fragmentation process and mechanisms of aggregation in association with the mean residential time and fate of plastics in aquatic environments
4) Assessment of rivers as the source of plastic pollutants to the oceans and estimation of flux of plastics from river environment to the open water system
5) Assessment and understanding of MPs interactions with living organisms followed by their impact on ecosystem services

Owing to their various sizes, resistance to degradation and ubiquitous distribution in the aquatic environment, plastics have been addressed as the great environmental concerns. In recent years, investigation of behavior of plastics and their toxicity with respect to interaction with pollutants and ingestion by biota have been started in aquatic ecosystems using field monitoring systems and environmental samples. The plastic particles are ingested by a variety of aquatic organisms from invertebrates to fish through various pathways. However, the presence and accumulation of unrecoverable plastics are a growing concern with many

unknown mechanisms and pathways in the aquatic environment. It is clear that larger plastics may cause choking, blocking of digestive tracts, false sense of satiation, starvation or death, while the impact on aquatic organisms of ingesting MP particles are largely unknown (van Weert et al., 2019). Since biological consequences of MPs ingestion by various aquatic organisms started to focus on understanding impact of MPs ingestion on the aquatic organisms and further recognition of the process on human health.

The assessment of terrestrial rivers to be a source of plastic particles to the oceans and estimation of flux and inputs of plastics from the river environment to the open water system are also considered as important research interest in this research field. The release of plastics into the marine environment occurs through a variety of pathways, including river flow, atmospheric transport, beach littering and directly to sea via aquaculture, shipping and fishing activities. Global monitoring of marine plastic debris succeeded to understand the fate, distribution and potential risks of plastic items, while behavior of plastic debris from its occurrence to release into marine environment in the terrestrial environment including river ecosystems is still largely unknown. The estimated amount of plastic debris from river to marine environment was from several to tens million tons of plastics fluxes to the ocean. However, the estimation based on large datasets instead of detailed investigation of river basin in the terrestrial environment loses important trends of plastic behavior in the riverine environment. In this case, regional studies are also necessary to identify plastic flux in the river system in order to evaluate input of plastic debris to the open water system.

6.4 A proposal for plastic research in terrestrial environment

At present, there is little understanding for the fate and transport of plastics particularly micro and nano-plastics not only in the Selenga River system but also other river environments across the globe due to inadequate investigation based on limited monitoring data. Information of environmental plastics in the long-term consequences is less addressed despite of various processes and mechanisms of plastics in the environment. This study will suggest the more general and region-specific monitoring and modeling of plastic debris in the river basin in order to track plastic debris distribution, accumulation and fate in the environment. For further important understandings of the environmental plastics, long-term monitoring and modeling of plastic debris in the river catchment are essential to provide a picture of the human impacts on the environment and plastic pollution problems.

As described in Chapter 3–5, environmental factors influenced various characteristics of plastics including their degraded and fragmented status in the river environment. These effects of environmental factors are also identified in other river environments in developing cities in the world (Lahens et al., 2018; Peng et al., 2018). As pointed out by Kiessling et al., (2019), hazardous litters including various types of plastic debris accumulated on the river shores without any control. Those distributed plastic litters on the river shores have potentials to represent mismanagement of plastic wastes. Several scientific reports highlighted mismanaged plastic wastes and demonstrated the pathways in the environment to reach aquatic systems with conceptual prediction (Jambeck et al., 2015; Lebreton and Andrady, 2019). However, country data provided to illustrate the mismanaged plastic wastes in the world demonstrates lack of detailed information regarding to the transportation of plastic debris in the unit aquatic system. Furthermore, available data were not always consistent due to different methodologies to collect data. These many concerns to estimate and understand the mismanaged plastic wastes, arising the necessity of long-term projections with suitable consistent methodologies.

In order to identify regional, national and global trends in plastic distribution, it would be desirable to conduct monitoring of plastic debris in the aquatic environment and to collect meta-data through field monitoring. However, one of major difficulties in datasets collected from the various researches was no standard protocols for procedures to collect plastic pieces from the environmental matrix. Hand-sorting and bulk sampling techniques from the shores and sediments of rivers in Chapter 2 have potential to solve the issue relating to the sampling approaches. Since hand sorting followed by the size determination and identifying the morphotypes directly by naked eyes or through photographs are one of the easiest and traditional methods, regional and national monitoring programs can adopt the techniques with other hydrological long-term monitoring programs. Larger sized plastics can be identified by naked eyes, while some microplastics requires further microscopic detection. Although MPs are a newly recognized type of environmental pollution in aquatic systems, no monitoring of these contaminants is conducted mostly due to the lack of routine quantification. Long-term monitoring of plastic debris in the river environment would be important goal to compile meta-data and understand mismanagement of plastic waste in order to find out further solution to reduce waste release accompanying with modeling of plastics in the environment.

As discussed in Chapter 4, behavior and reactivity of PSFs were relatively characteristic in the Tuul River. Furthermore, various types of MP distribution pathways and their

interaction with foamed plastics have been investigated. The abundance of plastic debris, their origin, fragmentation and degradation of plastic pieces should be differently considered in the rivers based on their geographical background (Mani et al., 2015). In this case, accompanying with the regional and national monitoring of plastic debris in the river catchments, modeling of plastic loads, mass budget, fragmentation rate and further prediction of abundance of micro and nano-plastics in the river environment are essential to conduct. From the modeling studies based on long-term monitoring, it is possible to obtain comprehensive properties of environmental plastics in terms of regional, national and global scale.

To date, modeling studies developed to evaluate MP dynamics in aquatic environments despite of less data about plastic debris in aquatic environments. In this challenge, there are several different model approaches mostly using ocean circulation models (Hardesty et al., 2017). Most of models for plastic debris in the aquatic environments mainly faced to difficulties to use empirical data due to less dataset. From the modeling approaches, it is possible to expect detail knowledge of processes of plastic contamination in a river system and extracted data could be comparable and can be applicable to other environments. Combined study of long-term monitoring and modeling of plastic debris in the river environment would be crucial for the fundamental understandings of plastic debris and the relationship between hydrodynamic parameters and distribution pattern of plastics in the natural aquatic system.

6.5 Prospect

With respect to abundance and distribution of plastic debris in Mongolia, it is still insufficient to explain comprehensive distribution and behavior of micro- and nano-plastics relating to their future migration in the river network of Selenga River basin. Interactions between hydrological processes and plastic polymers should be taken into account to evaluate the environmental risk of MP and nanoplastics released in the river ecosystems. From the degradation of PSF plastics (Fig. 6.2) with various values of carbonyl index and significant relationship between the different size fractions (Chapter 4), further degradation of MPs into the nanoplastics (<1 μm) would be predictable in the river environment. The amount of nanoplastics in the environment has not been studied yet, because there are no reliable methods for the determination, quantification and assessment (Shen et al., 2019). Due to considerably small particle sizes, micro- and nano-plastics are widely distributed in

the aquatic environment. The recorded minimum sizes of MPs in the river bottom sediment in the present study was 28.4 μm, whereas the size distribution of majority part of MPs in the sediment was in the range of 100 – 200 μm (Chapter 5). Because of the instrumental limitation to track the environmental nanoplastics by field survey, many researches focused laboratory scale measurements in order to identify the toxicity of nanoplastics in the aquatic ecosystem (Besseling et al., 2014; Shen et al., 2019).

Extension of the current thesis research can realize to establish a general model for plastic pollution particularly micro- and nano-plastic distribution. To identify the relationship between river hydrological processes and plastic distribution in the river system, it is necessary to understand impacts from the environmental (precipitation, air temperature, river flow rate, discharge etc.) and anthropogenic (land use changes) factors. For these reasons, river hydrodynamic models are available for the future research. Assessing the potential impacts of climate and land use change on water fluxes and sediment transport will be evaluated using Soil and Water Assessment Tool (SWAT) model (Bauwe et al., 2019; Wei et al., 2019). The application of SWAT model to dynamics of plastic debris approaches appropriate estimation of a pollution scale in a unit area. For the future challenge, the SWAT model will be selected since it is one of the most commonly used and well-supported hydrological models being able to assess the impact of varying land management practices on water resources and on water pollution (Jodar-Abellan et al., 2019; Wang et al., 2019c; Zhang et al., 2019).

An indirect calibration module is expected to estimate MP transport and storage. This indirect calibration is based on relationship (ratio) between MPs and suspended sediments in river water, or MPs in sediments and land surface erosion. Since Chapter 5 highlighted the aggregates of plastic and fine sediment, it supports possibilities to predict relationships between concentrations of suspended sediment and MPs. After the calculation process, the mathematical model describing the dynamics of MP can be established to predict the fate of MPs in riverine environments. The model requires members of meteorological information, land uses and hydrological behavior of sediments to simulate behavior of MPs as a function of geographical parameter and their interactions with suspended sediments. The model outcomes support the information of MPs flux and storage in the terrestrial freshwater environment and sustain the development of models in the terrestrial environment connecting to open water reservoirs. Ultimately, from the relationship between hydrological parameters (precipitation, air temperature, flow rate, discharge etc.) and plastic particles

distribution in the river environment, it is possible to account the controlling factor (environmental or anthropogenic) for plastic debris.

Furthermore, conventional plastics (i.e., polyethylene and polypropylene) strongly resist to biodegradation, leading to increases their adverse impacts to the aquatic environments. A large number of polyethylene and polystyrene plastics were recorded from the river shores of Mongolia, likewise other terrestrial river environments. Most of developing cities have similar issues regarding to the non-biodegradable environmental plastic pollution and the regulations to manage the released waste. From the various perspectives of environmental plastics, released waste from the high-populated areas necessary to be replaced by biodegradable plastics in order to improve their waste management. Since the sources of biodegradable plastics are insufficient to replace a vast amount of current plastic consumption (Hopewell et al., 2009) and their production is still limited on the global plastic demand, existing modern problems of plastic debris are necessary to deal well by wise management of MSW in the country level regulations.

We have considerable knowledge about many of the environmental hazards, and information on negative influences on human health, but many concerns and uncertainties are still left over. The associated issues with plastics including their interaction with other pollutants and vectoring them into various environments are necessary to consider in the aquatic and terrestrial environments. According to global plastic demand, the production continuously grows approximately 9 percent per annum (PlasticsEurope, 2017). Consequently, the quantity of plastics produced in this century should be controlled by many approaches to avoid great concerns in the environment. Additionally, the future research on MP and nanoplastic dynamics can be applied to guide its future monitoring and environmental evaluations. Development of the model for accumulation of MPs in rivers would be very useful for societies being responsible for monitoring and reporting the plastic pollution as well as organizations managing clean-up activities and remediation strategies. There are many uncertainties to understand transportation and accumulation mechanisms of MPs, and in the future study, some insights can be offered to understand mechanisms of MP and nanoplastic behavior in the urban river environment.

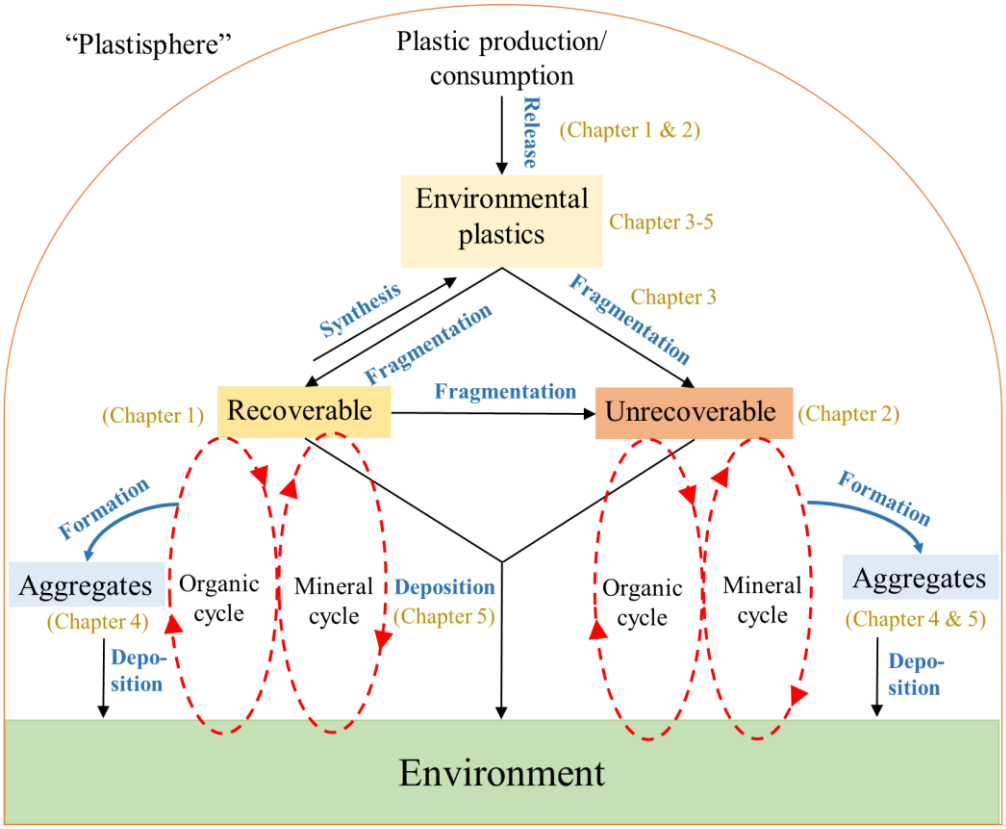

Fig. 6.1 Fate of environmental plastics.

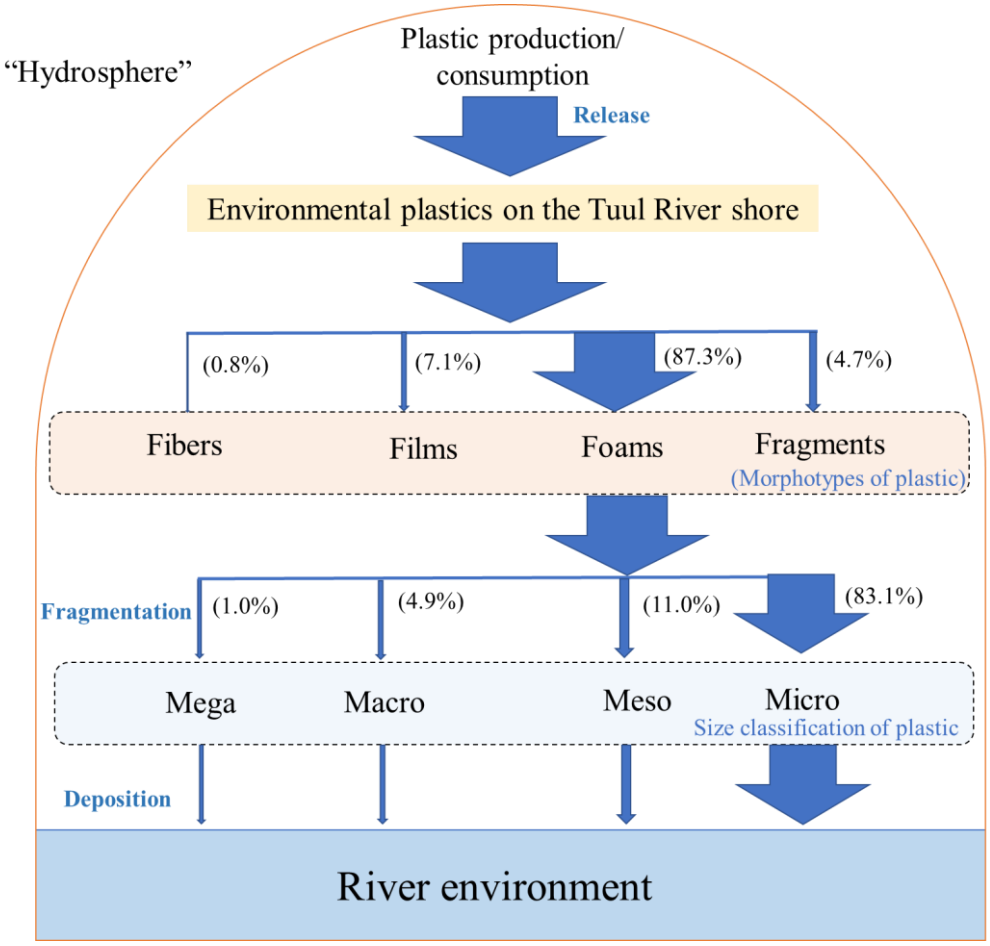

Fig. 6.2 Composition of environmental plastics and fragmentation rate of polystyrene foams (PSF) on the shore of Tuul River.

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